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(54) 【発明の名称】 R-T-B系焼結型永久磁石

(57) 【要約】

【課題】 高いBr及び高い(BH)maxが要求される用途に好適な高性能のR-T-B系焼結型永久磁石を提供する。

【解決手段】 重量百分率でR: 28~33%、B: 0.5~2%、及び残部実質的にT及び不可避免的不純物からなる組成を有するR-T-B系焼結型永久磁石 (RはYを含む少なくとも一種の希土類元素であって、Dy、Tb及びHoからなる群から選ばれた少なくとも一種の重希土類元素を必ず含み、TはFe又はFeとCoである。) であって、重希土類元素の濃度が結晶粒界相より高い第一のR<sub>2</sub>T<sub>14</sub>B型主相結晶粒と、前記重希土類元素の濃度が結晶粒界相より低い第二のR<sub>2</sub>T<sub>14</sub>B型主相結晶粒とを含有する組織を有するR-T-B系焼結型永久磁石。

## 【特許請求の範囲】

【請求項1】 重量百分率でR: 28～33%、B: 0.5～2%、及び残部実質的にT及び不可避的不純物からなる組成を有するR-T-B系統結型永久磁石(RはYを含む少なくとも一種の希土類元素であって、Dy、Tb及びHoからなる群から選ばれた少なくとも一種の重希土類元素を必ず含み、TはFe又はFeとCoである。)であって、前記重希土類元素の濃度が結晶粒相より高い第一の $R_2T_{14}B$ 型主相結晶粒と、前記重希土類元素の濃度が結晶粒相より低い第二の $R_2T_{14}B$ 型主相結晶粒とを含有する組織を有することを特徴とするR-T-B系統結型永久磁石。

【請求項2】 請求項1に記載のR-T-B系統結型永久磁石において、重量百分率で0.01～0.6%の $M_1$ (ただし $M_1$ はNb, Mo, W, V, Ta, Cr, Ti, Zr及びHfからなる群から選ばれた少なくとも一種の元素である。)を含有することを特徴とするR-T-B系統結型永久磁石。

【請求項3】 請求項1または2に記載のR-T-B系統結型永久磁石において、重量百分率で0.01～0.3%の $M_2$ (ただし $M_2$ はAl, Ga及びCuからなる群から選ばれた少なくとも一種の元素である。)を含有することを特徴とするR-T-B系統結型永久磁石。

【請求項4】 請求項1～3のいずれかに記載のR-T-B系統結型永久磁石において、重量百分率でRが31%を超えて33%以下であることを特徴とするR-T-B系統結型永久磁石。

【請求項5】 請求項1～4のいずれかに記載のR-T-B系統結型永久磁石において、前記不可避的不純物として、重量百分率で0.6%以下の酸素、0.15%以下の炭素、0.15%以下の窒素、及び0.3%以下のCaを含有することを特徴とするR-T-B系統結型永久磁石。

【請求項6】 請求項1～4のいずれかに記載のR-T-B系統結型永久磁石において、前記不可避的不純物として重量百分率で0.25%以下の酸素、0.15%以下の炭素、0.03%以下の窒素、及び0.3%以下のCaを含有することを特徴とするR-T-B系統結型永久磁石。

## 【発明の詳細な説明】

## 【0001】

【発明の属する技術分野】本発明は高い保磁力、残留磁束密度及び最大エネルギー積を有するR-T-B系統結型永久磁石に関する。

## 【0002】

【従来の技術及び発明が解決しようとする課題】R-T-B系統結型永久磁石(RはYを含む少なくとも一種の希土類元素であり、TはFe又はFeとCoである。)は、最大エネルギー積ではほぼ40MGOeのものが量産されている。R-T-B系統結型永久磁石の合金組成を調整する手段として、シングル法とブレンド法とがある。

【0003】シングル法は、溶解/鋳造段階でR-T-B系統結型永久磁石の主成分組成に調整したインゴットを用いて、粉碎、磁界中成形、焼結及び熱処理を行いR-T-B系

焼結型永久磁石を製造する方法であり、得られたR-T-B系統結型永久磁石は所望の機械加工及び表面処理を施して実用に供される。

【0004】ブレンド法は、組成の異なる2種以上のR-T-B系統結型永久磁石用合金粉末を最終的に所望のR-T-B系統結型永久磁石の主成分組成になる配合比で混合した後、必要に応じて粉碎を行い、以後磁界中成形、焼結、熱処理及び表面処理を行いR-T-B系統結型永久磁石を製造する方法である。

【0005】上記シングル法によれば、保磁力*iH<sub>c</sub>*を高めるのは比較的容易であるが、残留磁束密度*B<sub>r</sub>*及び最大エネルギー積(*BH*)*max*が低くなり、高い*B<sub>r</sub>*及び高い(*BH*)*max*が要求される用途には適さないという問題がある。

【0006】また従来のブレンド法の適用例として、R含有量の高いR-T系合金とR含有量の低いR-T-B系合金とを配合してなるR-T-B系統結型永久磁石(特開平7-122413号)や、Ga, C, OがRリッチ相及びその周辺に偏析しているR-T-B系統結型永久磁石(特開平9-232121号)が提案されているが、高い*B<sub>r</sub>*及び高い(*BH*)*max*の用途に好適にするに改良の余地がある。特に磁気特性への影響が大きい主相結晶粒の重希土類元素の最適な濃度分布及びその制御方法については、何も説明されていない。

【0007】従って本発明の目的は、高い*B<sub>r</sub>*及び高い(*BH*)*max*が要求される用途に好適な高性能のR-T-B系統結型永久磁石を提供することである。

## 【0008】

【課題を解決するための手段】上記目的に鑑み鋭意研究の結果、本発明者らは、重量百分率でR: 28～33%、B: 0.5～2%、残部実質的にT及び不可避的不純物からなる組成(RはYを含む少なくとも一種の希土類元素であって、Dy、Tb及びHoからなる群から選ばれた少なくとも一種の重希土類元素を必ず含み、TはFe又はFeとCoである。)を有し、前記重希土類元素の濃度が結晶粒相より高い第一の $R_2T_{14}B$ 型主相結晶粒と、前記重希土類元素の濃度が結晶粒相より低い第二の $R_2T_{14}B$ 型主相結晶粒とを含有する組織を有するR-T-B系統結型永久磁石が、高い*B<sub>r</sub>*及び高い(*BH*)*max*を示すことを見出し本発明に想到した。

【0009】本発明の好ましい一実施例によるR-T-B系統結型永久磁石は、重量百分率でR: 28～33%、B: 0.5～2%、 $M_1$ : 0.01～0.6% ( $M_1$ はNb, Mo, W, V, Ta, Cr, Ti, Zr及びHfからなる群から選ばれた少なくとも一種の元素である。)、残部実質的にT及び不可避的不純物からなる組成を有する。

【0010】本発明の好ましい別の実施例によるR-T-B系統結型永久磁石は、重量百分率でR: 28～33%、B: 0.5～2%、 $M_1$ : 0.01～0.6% ( $M_1$ はNb, Mo, W, V, Ta, Cr, Ti, Zr及びHfからなる群から選ばれた少なくとも一種の元素である。)、 $M_2$ : 0.01～0.3% ( $M_2$ はAl, Ga及びCuからなる群から選ばれた少なくとも一種の元素であ

る。)、残部実質的にT及び不可避的不純物からなる組成を有する。

【0011】本発明のさらに好ましい実施例によるR-T-B系統結型永久磁石は、重量百分率でRが31%を超えて33%以下であり、かつ不可避的不純物として0.6%以下の酸素、0.15%以下の炭素、0.03%以下の窒素、及び0.3%以下のCaを含有する組成を有する。

【0012】本発明のさらに好ましい実施例によるR-T-B系統結型永久磁石は、重量百分率でRが28~31%であり、かつ不可避的不純物として0.25%以下の酸素、0.15%以下の炭素、0.15%以下の窒素、及び0.3%以下のCaを含有する組成を有する。

【0013】本発明のR-T-B系統結型永久磁石は、例えば希土類元素の全量は同じで、重希土類元素(Dy等)/軽希土類元素(Nd, Pr等)の比率が異なる以外は組成が実質的に同じ2種類以上の合金粉末を混合し、磁界中成形、焼結及び熱処理を行い、次いで必要に応じて機械加工、仕上げ加工(バレル加工等)、及び表面処理(Niめっき等)を行なうことより得られる。前記2種類以上の合金粉末の組成及びR-T-B系統結型永久磁石の最終組成に応じて、最適の焼結条件を選択し、もって焼結体組織内での重希土類元素(Dy等)の拡散状態を厳密に制御するのが重要である。その結果、 $R_2T_{14}$ B型主相結晶粒(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy等)の濃度分布に関して、重希土類元素(Dy等)の濃度が結晶粒界相より高い $R_2T_{14}$ B型主相結晶粒と、重希土類元素(Dy等)の濃度が結晶粒界相より低い $R_2T_{14}$ B型主相結晶粒とを含有する結晶組織が得られる。

【0014】このような焼結体組織を有するR-T-B系統結型永久磁石は、シングル法によるR-T-B系統結型永久磁石に比べて保磁力*iHc*はやや低いが、格段に高いBr及び(BH)<sub>max</sub>を有する。これと重希土類元素(Dy等)の濃度分布との相関はまだ十分に明確になっていないが、重希土類元素(Dy等)濃度が結晶粒界相より高い $R_2T_{14}$ B型主相結晶粒が高いBrの実現に寄与し、重希土類元素(Dy等)濃度が結晶粒界相より低い $R_2T_{14}$ B型主相結晶粒がシングル法に近い高い*iHc*の実現に寄与しているものと推定される。

【0015】

【発明の実施の形態】[1] R-T-B系統結型永久磁石

(A) 組成

(a) 主成分

本発明のR-T-B系統結型永久磁石の組成は、重量百分率でR 28~33%、B 0.5~2%及びTからなる主成分と不可避的不純物とからなる。その他に、主成分として、0.01~0.6重量%の $M_1$ (Nb, Mo, W, V, Ta, Cr, Ti, Zr及びHfからなる群から選ばれた少なくとも一種の元素)及び/又は0.01~0.3重量%の $M_2$ (Al, Ga及びCuからなる群から選ばれた少なくとも一種の元素)を含有するのが好ましい。

【0016】(1) R元素

R元素はYを含む少なくとも一種の希土類元素であって、Dy, Tb及びHoからなる群から選ばれた少なくとも一種の重希土類元素を必ず含む。重希土類元素以外の希土類元素(Yを含む)としては、Nd, Pr, La, Sm, Ce, Eu, Gd, Er, Tm, Yb, Lu及びYが挙げられる。希土類元素Rとして、ミッシュメタルやジジムのよう二種以上の希土類元素の混合物を用いてもよい。

【0017】Rの含有量は28~33重量%である。Rの含有量が28重量%未満であると、実用に耐える高い*iHc*が得られず、また33重量%を超えるとBrの低下が顕著になる。

【0018】重希土類元素の含有量は、0.2~15重量%の範囲内であるのが好ましい。重希土類元素の含有量が0.2重量%未満であると、結晶組織中の重希土類元素の分布による磁気特性の向上効果が不十分である。また重希土類元素の含有量が15重量%を超えると、R-T-B系統結型永久磁石のBr及び(BH)<sub>max</sub>は大きく低下する。より好ましい重希土類元素の含有量は0.5~13重量%である。

【0019】(2) B

Bの含有量は0.5~2重量%である。Bの含有量が0.5重量%未満であると、実用に耐える高い*iHc*を得るのが困難であり、また2重量%を超えるとBrの低下が顕著になる。

【0020】(3) T元素

T元素はFe単独又はFe+Coである。Coの添加により焼結型永久磁石の耐食性が改善されるとともに、キュリー一点が上昇して永久磁石としての耐熱性が向上する。しかしCoの含有量が5重量%を超えるとR-T-B系統結型永久磁石の磁気特性に有害なFe-Co相が形成され、Brと*iHc*がともに低下する。従ってCo含有量は5重量%以下とする。一方、Co含有量が0.5重量%未満では耐食性の改善効果や耐熱性の向上効果が不十分である。従ってCoを添加する場合、Co含有量を0.5~5重量%とするのが好ましい。

【0021】(4)  $M_1$  元素

$M_1$ はNb, Mo, W, V, Ta, Cr, Ti, Zr及びHfからなる群から選ばれた少なくとも一種の高融点金属元素である。 $M_1$ 元素の存在により、焼結過程において重希土類元素(Dy等)の拡散によって生じる主相結晶粒の過度の成長が抑えられ、シングル法に近い高い*iHc*を安定して得ることができる。しかし、 $M_1$ 元素を過剰に添加すると、逆に主相結晶粒の正常な粒成長が阻害され、Brの低下を招く。よって、 $M_1$ 元素の含有量の上限は0.6重量%である。一方、 $M_1$ 元素の含有量が0.01重量%未満では十分な添加効果が認められない。従って、 $M_1$ 元素の含有量は0.01~0.6重量%であるのが好ましい。

【0022】(5)  $M_2$  元素

$M_2$ はAl, Ga及びCuからなる群から選ばれた少なくとも一

種の元素である。

【0023】Alの微量添加によってR-T-B系統結型永久磁石のiHc及び耐食性が改善される。しかしAl含有量が0.3重量%を超えるとBrが大きく低下するのでAl含有量は0.3重量%以下とする。一方、Al含有量が0.01重量%未満ではiHcや耐食性の改善効果が不十分である。

【0024】Gaの微量添加によりR-T-B系統結型永久磁石のiHcが顕著に向上する。しかしAlと同様に、0.3重量%を超えるとBrが大幅に低下するのでGa含有量は0.3重量%以下とする。また0.01重量%未満ではiHcの意味ある向上効果が認められない。

【0025】Cuの微量添加は焼結体の耐食性の改善とiHcの向上に有効である。しかしAl及びGaと同様、Cu含有量が0.3重量%を超えるとR-T-B系統結型永久磁石のBrが大幅に低下し、また0.01%未満では耐食性の改善及びiHcの向上効果が不十分である。

【0026】以上の通り、 $M_2$ 元素の含有量は、Al、Ga又はCuのいずれも0.01~0.3重量%とする。

【0027】(b) 不可避的不純物

不可避的不純物としては、酸素、炭素、窒素、カルシウム等が挙げられる。Caは重希土類元素の含有量が異なる二種類以上のR-T-B系合金を還元拡散法(希土類元素の酸化物粉末を還元剤(Ca)で還元し、続いて他の主成分金属との相互拡散により合金粉末を得る方法)により作製した場合に、不可避的不純物として混入する。

【0028】酸素の含有量は0.6重量%以下であるのが好ましく、炭素の含有量は0.15重量%以下であるのが好ましく、窒素の含有量は0.15重量%以下であるのが好ましく、カルシウムの含有量は0.3重量%以下であるのが好ましい。各不可避的不純物の含有量が上記上限を超えると、R-T-B系統結型永久磁石の磁気特性は低下する。より好ましい不可避的不純物の含有量としては、酸素が0.25重量%以下であり、炭素が0.15重量%以下であり、窒素が0.03重量%以下である。特に好ましい不可避的不純物の含有量は、酸素が0.05~0.25重量%であり、炭素が0.01~0.03%であり、窒素が0.02~0.15%である。

【0029】このような不可避的不純物量を有するR-T-B系統結型永久磁石の組成の具体例としては、以下のものが挙げられる。

【0030】(i) 重量百分率でRが31%を超えて33%以下であり、酸素が0.6%以下であり、炭素が0.15%以下であり、窒素が0.03%以下であり、Caが0.3%以下である組成。例えば乾式成形法の採用により、酸素を0.25~0.6%、炭素を0.01~0.15%、窒素を0.005~0.03%にすることができる。

【0031】(ii) 重量百分率でRが28~31%であり、酸素が0.25%以下であり、炭素が0.15%以下であり、窒素が0.15%以下であり、Caが0.3%以下である組成。例えば湿式成形法の採用により、酸素を0.05~0.25重量%、炭素を0.01~0.15%、窒素を0.02~0.15%にすることが

できる。

【0032】(B) 組織

本発明のR-T-B系統結型永久磁石の結晶組織は $R_2T_{14}B$ 型主相結晶粒と結晶粒界相を有し、前記 $R_2T_{14}B$ 型主相結晶粒は少なくとも、(i)重希土類元素の濃度が結晶粒界相より高い第一の $R_2T_{14}B$ 型主相結晶粒と、(ii)重希土類元素の濃度が結晶粒界相より低い第二の $R_2T_{14}B$ 型主相結晶粒とを含有する。前記 $R_2T_{14}B$ 型主相結晶粒はさらに(iii)重希土類元素の濃度が結晶粒界相とはほぼ等しい第三の主相結晶粒を含有しても良い。ここで $R_2T_{14}B$ 型主相結晶粒における重希土類元素の濃度は $R_2T_{14}B$ 型主相結晶粒のほぼ中心部(芯部)において測定したものであり、 $R_2T_{14}B$ 型主相結晶粒の芯部とは結晶粒界から1.0 $\mu$ m以上入り込んだ領域をいう。重希土類元素としてはDyが好ましいが、Tb及び/又はHo、又はそれらとDyとの混合物であっても良い。

【0033】撮影した結晶組織の断面写真において $R_2T_{14}B$ 型主相結晶粒の総個数を100%として、第一の $R_2T_{14}B$ 型主相結晶粒の個数の比率は1~35%であり、第二の $R_2T_{14}B$ 型主相結晶粒の個数の比率は3~55%であり、第三の $R_2T_{14}B$ 型主相結晶粒の個数の比率は96~10%であるのが好ましい。第一~第三の $R_2T_{14}B$ 型主相結晶粒の個数の比率が上記範囲外であると、R-T-B系統結型永久磁石は高い保磁力iHc、残留磁束密度Br及び最大エネルギー積(BH)<sub>max</sub>を有するのが困難である。より好ましくは、第一の $R_2T_{14}B$ 型主相結晶粒の個数の比率が3~30%であり、第二の $R_2T_{14}B$ 型主相結晶粒の個数の比率が10~45%であり、第三の $R_2T_{14}B$ 型主相結晶粒の個数の比率が87~25%である。

【0034】(2) 製造方法

上記組織を有する本発明のR-T-B系統結型永久磁石を製造するには、例えばDy等の重希土類元素の含有量が異なる二種類以上のR-T-B系合金粉末を混合するいわゆるブレンド法を採用する。この場合、各R-T-B系合金粉末の組成は、R元素の合計量が各合金粉末で同じになるようにする。例えばNd+Dyの場合、後述の実施例1に示すように、一方の合金粉末を29.0%Nd+1.0%Dyとし、他方の合金粉末を15.0%Nd+15.0%Dyとする。R元素以外の元素については、各合金粉末が実質的に同じであるのが好ましいが、 $M_1$ 及び/又は $M_2$ の含有量に若干の差があっても良い。

【0035】例えば二種類の合金粉末を混合する場合、両者のR元素の合計量を同じにするとともに、第一の合金粉末中における重希土類元素の含有量を0~10重量%とし、第二の合金粉末中における重希土類元素の含有量を10重量%超40重量%以下とする。この場合、第一の合金粉末/第二の合金粉末の配合比を重量で70/30~95/5とするのが好ましく、80/20~90/10とするのがより好ましい。これは、第一の合金粉末と第二の合金粉末との間の重希土類元素の含有量の差が大きくなるほど、第

一の合金粉末と第二の合金粉末との間の微粉碎性（微粉の粒径分布）の差異が大きくなり、最終的に得られるR-T-B系統焼結型永久磁石の主相結晶粒の粒径分布が幅広くなり、磁化の強さ（ $4\pi I$ ）-磁界の強さ（H）の関係を示す減磁曲線の角形性及び（BH）maxの劣化を招来するからである。

【0036】R-T-B系合金粉末の微粉碎は、不活性ガスを媒体とするジェットミル等の乾式粉碎法又はボールミル等の湿式粉碎法により行うことができる。高い磁気特性を得るために、実質的に酸素を含有しない（濃度：体積比で1000ppm以下）不活性ガス雰囲気中でジェットミル微粉碎後、大気に触れないようにして不活性ガス雰囲気中から微粉を直接鉱油、合成油、植物油又はそれらの混合油中に回収し、混合物（スラリー）にするのが好ましい。微粉を大気から遮断することにより、酸化及び水分の吸着を抑制することができる。鉱油、合成油又は植物油として、脱油性及び成形性の観点から、分留点が350℃以下のものが好ましく、動粘度は室温において10cSt以下のものがよく、5cSt以下のものがより好ましい。

【0037】本発明の永久磁石を製造するために配合する2種以上のR-T-B系統焼結型永久磁石用合金として、日本国特許第2,665,590号、日本国特許第2,745,042号等に例示されている薄板状合金（ストリップキャスト合金）を使用してもよい。この薄板状合金（ストリップキャスト合金）は、本発明の要件を満たす組成を有する合金溶湯を単ロール法、双ロール法又は回転ディスク法等の溶湯急冷法により急冷、凝固してなり、ほぼ柱状結晶の均質な組織を有し、かつ前記柱状結晶の短軸方向の平均結晶粒径が3～20 $\mu$ mである。高いBr及び（BH）maxを得るためには、薄板状合金を不活性ガス（Ar等）雰囲気中で900～1200℃×1～10時間加熱後室温まで冷却する均質化熱処理を行った後、粉碎するのが好ましい。

【0038】混合物（スラリー）を用いて、所望の成形装置により磁界中で湿式成形することにより成形体を得る。酸化による磁気特性の劣化を抑えるために、成形直後から焼結炉に入れるまでの間、油中又は不活性ガス雰

囲気中に保持するのが望ましい。成形は乾式法により行なっても良い。乾式成形法の場合、不活性ガス雰囲気中で乾燥微粉の混合物を磁場中でプレス成形する。

【0039】湿式成形体の焼結に際し、常温から焼結温度まで急激に昇温すると、成形体中に残留した鉱油、合成油又は植物油が希土類元素と反応して希土類炭化物を生成し、得られる焼結磁石の磁気特性の劣化を招く。この対策として、温度100～500℃、真空度 $10^{-1}$  Torr以下で30分以上保持する脱油処理を施すのが望ましい。脱油処理により成形体中に残留する鉱油、合成油又は植物油を十分に除去することができる。なお加熱温度は100～500℃の温度範囲であれば一定である必要はない。また $10^{-1}$  Torr以下の真空度で室温から500℃まで昇温する間、昇温速度を10℃/分以下、好ましくは5℃/分以下としても、ほぼ同等の脱油効果を得ることができる。

【0040】成形体を不活性ガス雰囲気中で約1000～1200℃の温度で焼結することにより、R-T-B系統焼結型永久磁石を製造する。得られたR-T-B系統焼結型永久磁石に所望の機械加工及び表面処理を施す。表面処理としては、Niめっきや電着エポキシ樹脂コーティング等が挙げられる。

#### 【0041】

【実施例】本発明を以下の実施例によりさらに詳細に説明するが、本発明はそれらに限定されるものではない。

#### 【0042】実施例1

表1の主成分組成を有する溶製合金A及び溶製合金Bをそれぞれ不活性ガス雰囲気中で粗粉碎し、篩分することにより粒径500 $\mu$ m以下の粗粉を得た。合金Aの粗粉87.9kgと合金Bの粗粉12.1kgとをV型混合機に投入して混合し、100kgの混合粗粉を得た。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd27.3%、Dy2.7%、B1.0%、Nb0.2%、Al0.1%、Co1.0%、Cu0.1%、残部Feであり、この混合粗粉に含有される不純物は、0.15重量%のO、0.01重量%のN、及び0.02重量%のCであった。

#### 【0043】

【表1】

合金	組成（重量%）						
	Nd	Dy	B	Nb	Al	Co	Cu
A	29.0	1.0	1.0	0.2	0.1	1.0	0.1
B	15.0	15.0	1.0	0.2	0.1	1.0	0.1

【0044】前記混合粗粉を酸素濃度が10ppm以下（体積比）の窒素ガス雰囲気中でジェットミル粉碎し、平均粒径4.0 $\mu$ mの微粉とした。微粉を窒素ガス雰囲気中で大気と非接触状態で直接鉱油（出光興産（株）製、商品名：出光スーパーゾルPA-30）中に回収し、微粉スラリーを得た。この微粉スラリーを用いて磁界強度10kOe及び成形圧1.0ton/cm<sup>2</sup>の条件で湿式圧縮成形し、得られた成形体を約 $5 \times 10^{-1}$  Torrの真空中で200℃で1時間加熱して脱油後、引き続き約 $3 \times 10^{-5}$  Torrで1050～1100℃

の温度範囲で各々2時間焼結し、室温まで冷却して焼結体を得た。

【0045】各焼結体に不活性ガス雰囲気中で900℃×2時間と500℃×1時間の熱処理を各1回施した後、室温まで冷却してR-T-B系統焼結型永久磁石を得た。20℃において磁気特性を測定したところ、図1に示す結果を得た。図1から明らかなように、焼結温度を1070～1110℃とした場合に永久磁石として好ましい磁気特性が得られた。特に焼結温度を1090℃とした場合に13.8kGのBr、18

kOeのiHc及び45.9MGOeの(BH)maxが得られ、焼結温度を100℃とした場合に13.8kGのBr、17.9kOeのiHc、45.7MGOeの(BH)maxが得られ、Br及び(BH)maxが高かった。

【0046】前記焼結磁石のうち代表的な焼結磁石の組成を分析したところ、重量百分率で主成分はNd: 27.3%、Dy: 2.7%、B: 1.0%、Nb: 0.2%、Al: 0.1%、Co: 1.0%、Cu: 0.1%、残部: Feであり、不可避的不純物は、0.17%のO、0.05%のN、及び0.07%のCであった。

【0047】前記焼結磁石のうち代表的な焼結磁石の断面組織を後述の実施例7と同様にして観察し、主相結晶粒( $R_2T_{14}B$ )内(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy)の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy)の濃度が結晶粒界相

より高い第一の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相より低い第二の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相とほぼ等しい第三の主相結晶粒とから構成されていることが分かった。

#### 【0048】比較例1

表2の主成分組成を有する溶製合金Cを用いた以外は実施例1と同様にして粗粉碎した。この粗粉の組成(重量%)を分析したところ、主成分はNd: 27.3%、Dy: 2.7%、B: 1.0%、Nb: 0.2%、Al: 0.1%、Co: 1.0%、Cu: 0.1%、残部: Feであり、不純物はO: 0.13%、N: 0.008%、C: 0.02%であった。

#### 【0049】

#### 【表2】

合金	組成(重量%)							
	Nd	Dy	B	Nb	Al	Co	Cu	Fe
C	27.3	2.7	1.0	0.2	0.1	1.0	0.1	殘部

【0050】この粗粉を用いて実施例1と同様にして微粉碎(平均粒径 $4.1\mu m$ )、スラリー化、磁界中成形、脱油、焼結及び熱処理を行い、シングル法による比較例の焼結型永久磁石を得た。この焼結型永久磁石の組成(重量%)を分析したところ、主成分は、Nd: 27.3%、Dy: 2.7%、B: 1.0%、Nb: 0.2%、Al: 0.1%、Co: 1.0%、Cu: 0.1%、残部: Feであり、不純物は、O: 0.15%、N: 0.04%、C: 0.06%であった。

【0051】20℃において磁気特性を測定した結果を図1に示す。図1より、iHcのレベルは19kOe前後と高いが、Brは13.3kG以下、(BH)maxは42.5MGOe以下であり、実施例1のBr及び(BH)maxに比べて低いことが分かる。またこの比較例の焼結磁石の断面組織には、重希土類元

素Dyの濃度が結晶粒界相より高い主相結晶粒は観察されなかった。

#### 【0052】実施例2

表3の主成分組成を有する溶製合金D及び溶製合金Eをそれぞれ用いた以外は実施例1と同様にして粗粉碎した。合金Dの粗粉94kgと合金Eの粗粉6kgとをV型混合機に投入して混合し、100kgの混合粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分はNd: 22.4%、Pr: 8.9%、Dy: 1.2%、B: 1.0%、Al: 0.1%、Ga: 0.15%、残部: Feであり、不純物はO: 0.14%、N: 0.01%、C: 0.01%であった。

#### 【0053】

#### 【表3】

合金	組成(重量%)						
	Nd	Pr	Dy	B	Al	Ga	Fe
D	23.2	9.3	—	1.0	0.1	0.15	残部
E	8.9	3.6	20.0	1.0	0.1	0.15	残部

【0054】混合粗粉を酸素濃度が500ppm(体積比)以下の窒素ガス雰囲気中でジェットミル粉碎し、平均粒径 $4.1\mu m$ の微粉とした。この微粉を用いて磁界強度10kOe、成形圧 $1.5\text{ton}/\text{cm}^2$ の条件で乾式圧縮成形した。得られた成形体を約 $3 \times 10^{-5}$  Torrの真空中、1040~1110℃の温度範囲で各々2時間焼結した後、室温まで冷却して焼結体を得た。

【0055】各焼結体に不活性ガス雰囲気中で900℃×3時間と550℃×1時間の熱処理を各1回施した後、室温まで冷却してR-T-B系焼結型永久磁石を得た。20℃で磁気特性を測定したところ、図2に示す結果を得た。図2から、焼結温度を1050~1100℃とした場合に永久磁石として好ましい磁気特性が得られることが分かった。特に焼結温度が1070℃の場合に13.4kGのBr、16.3kOeのiHc、及び43.2MGOeの(BH)maxが得られ、また焼結温度が10

80℃の場合に13.4kGのBr、15.1kOeのiHc、及び43.3MGOeの(BH)maxが得られ、Br及び(BH)maxが高かった。

【0056】前記焼結磁石のうち代表的な焼結磁石の組成を分析したところ、重量百分率で主成分はNd: 22.4%、Pr: 8.9%、Dy: 1.2%、B: 1.0%、Al: 0.1%、Ga: 0.15%、残部: Feであり、不純物はO: 0.45%、N: 0.02%、C: 0.07%であった。

【0057】前記焼結磁石のうち代表的な焼結磁石の断面組織を後述の実施例7と同様にして主相結晶粒( $R_2T_{14}B$ )内(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy)の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy)の濃度が結晶粒界相より高い第一の $R_2T_{14}B$ 型主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相より低い第二の $R_2T_{14}B$ 型主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相とほぼ等しい第三

の $R_2T_{14}B$ 型主相結晶粒とから構成されていることが分かった。

#### 【0058】比較例2

表4の主成分組成を有する溶製合金Fを用いた以外は実施例1と同様にして粗粉碎した。粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 22.4%, Pr: 8.9

%, Dy: 1.2%, B: 1.0%, Al: 0.1%, Ga: 0.15%, 残部: Feであり、不純物はO: 0.14%, N: 0.01%, C: 0.02%であった。

#### 【0059】

【表4】

合金	組成 (重量%)						
	Nd	Pr	Dy	B	Al	Ga	Fe
F	22.4	8.9	1.2	1.0	0.1	0.15	残部

【0060】この粗粉を用いて実施例2と同様にして微粉碎（平均粒径 $4.0\mu m$ ）、磁界中成形、焼結及び熱処理を行い、シングル法による比較例の焼結型永久磁石を得た。この磁石の成分を分析したところ、重量百分率で主成分はNd: 22.4%, Pr: 8.9%, Dy: 1.2%, B: 1.0%, Al: 0.1%, Ga: 0.15%, 残部: Feであり、不純物はO: 0.43%, N: 0.03%, C: 0.06%であった。

【0061】20℃で磁気特性を測定した結果を図2に示す。図2から明らかなように、 $iH_c$ のレベルは実施例2に比べてやや高いが、 $Br_i$ は12.9kG以下で、 $(BH)_{max}$ は40.1MGOe以下と低かった。またこの比較例の焼結磁石の断面組織には、重希土類元素 (Dy) の濃度が結晶粒界相より高い主相結晶粒は観察されなかった。

#### 【0062】実施例3

表5の主成分組成を有する溶製合金G及び溶製合金Hを用いた以外は実施例1と同様にして粗粉碎した。次に合金Gの粗粉81.8kgと合金Hの粗粉18.2kgとをV型混合機に投入して混合し、100kgの混合粗粉を得た。混合粗粉の組成を分析したところ、重量百分率で主成分はNd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.29%, Al: 0.10%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部: Feであり、不純物はO: 0.14%, N: 0.01%, C: 0.02%であった。

#### 【0063】

【表5】

合金	組成 (%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
G	22.29	6.21	2.00	0.97	0.35	0.10	2.00	0.08	0.10	残部
H	5.03	1.47	24.00	0.97	—	0.10	2.00	0.08	0.10	残部

【0064】この混合粗粉を用いて実施例1と同様にして微粉碎（平均粒径 $4.2\mu m$ ）、スラリー化、磁界中圧縮成形を行った。得られた成形体を約 $5 \times 10^{-1}$  Torrの真空中で200℃で1時間加熱して脱油し、次いで約 $2 \times 10^{-5}$  Torrの真空中、1060～1130℃の温度範囲内の各温度で2時間焼結後、室温まで冷却した。得られた各焼結体に不活性ガス雰囲気中で900℃×2時間と500℃×1時間の熱処理を各1回施した後、室温まで冷却してR-T-B系焼結型永久磁石を得た。20℃において磁気特性を測定した結果を図3に示す。図3から明らかなように、焼結温度を1070～1120℃とした場合に永久磁石として好ましい磁気特性が得られた。特に焼結温度を1100℃とした場合に12.7kGの $Br$ 、25.5kOeの $iH_c$ 及び38.8MGOeの $(BH)_{max}$ が得られ、1110℃とした場合に12.7kGの $Br$ 、25.3kOeの $iH_c$ 及び38.6MGOeの $(BH)_{max}$ が得られ、 $Br$ 及び $(BH)_{max}$ が高かった。

【0065】前記永久磁石のうち代表的な永久磁石の組成を分析したところ、重量百分率で主成分はNd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.29%, Al: 0.10%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部Feであり、不純物はO: 0.16%, N: 0.05%, C: 0.

07%であった。

【0066】焼結温度1100℃及び1110℃の条件で作製した前記永久磁石の断面組織について、後述の実施例7と同様にして主相結晶粒 ( $R_2T_{14}B$ ) 内 (ほぼ中心部) 及び結晶粒界相における重希土類元素 (Dy) の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素 (Dy) の濃度が結晶粒界相より高い第一の主相結晶粒と、重希土類元素 (Dy) の濃度が結晶粒界相より低い第二の主相結晶粒と、重希土類元素 (Dy) の濃度が結晶粒界相とほぼ等しい第三の主相結晶粒とから構成されていることが分かった。

#### 【0067】比較例3

表6の主成分組成を有する溶製合金Iを用いた以外は実施例1と同様にして粗粉を得た。この粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.29%, Al: 0.10%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部: Feであり、不純物はO: 0.12%, N: 0.01%, C: 0.01%であった。

#### 【0068】

【表6】

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
I	19.14	5.34	6.00	0.97	0.29	0.10	2.00	0.08	0.10	残部

【0069】この粗粉を用いた以外は実施例1と同様にして微粉砕（平均粒径 $4.2\mu\text{m}$ ）、スラリー化及び磁界中成形を行った。得られた成形体に対し、実施例3と同条件で脱油、焼結及び熱処理を行い、シングル法による比較例の焼結型永久磁石を得た。この磁石の組成を分析したところ、重量百分率で主成分は、Nd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.29%, Al: 0.10%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部: Feであり、不純物はO: 0.14%, N: 0.04%, C: 0.06%であった。

【0070】 $20^\circ\text{C}$ で磁気特性を測定した結果を図3に示す。図3から明らかなように、 $iH_c$ のレベルは $25\text{kOe}$ 前後と高いが、 $Br$ は $12.2\text{kG}$ 以下、 $(BH)_{\text{max}}$ は $35.7\text{MGoe}$ 以下であり、実施例3に比べて低かった。またこの比較例の焼

結磁石の断面組織において、重希土類元素（Dy）の濃度が結晶粒界相より高い主相結晶粒は観察されなかった。

#### 【0071】比較例4

表7の主成分組成を有する溶製合金J及び溶製合金Kをそれぞれ用いた以外は実施例1と同様にして粗粉砕した。合金Jの粗粉 $81.8\text{kg}$ と合金Kの粗粉 $18.2\text{kg}$ とをV型混合機に投入して混合し、 $100\text{kg}$ の混合粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.65%, Al: 0.10%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部: Feであり、不純物はO: 0.15%, N: 0.02%, C: 0.02%であった。

#### 【0072】

【表7】

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
J	22.29	6.21	2.00	0.97	0.80	0.10	2.00	0.08	0.10	残部
K	5.03	1.47	24.00	0.97	-	0.10	2.00	0.08	0.10	残部

【0073】この粗粉を用いた以外は実施例1と同様にして微粉砕（平均粒径 $4.1\mu\text{m}$ ）、スラリー化及び磁界中成形を行った。得られた成形体を約 $5 \times 10^{-1}$  Torrの真空中で $200^\circ\text{C}$ で1時間加熱して脱油し、次いで約 $2 \times 10^{-5}$  Torrの真空中で $1060 \sim 1130^\circ\text{C}$ の温度範囲内の各温度で2時間焼結後、室温まで冷却した。得られた各焼結体に不活性ガス雰囲気中で $900^\circ\text{C} \times 2$ 時間と $500^\circ\text{C} \times 1$ 時間の熱処理を各1回施した後、室温まで冷却して、ブレンド法による比較例の焼結型永久磁石を得た。 $20^\circ\text{C}$ において磁気特性を測定した結果を図3に示す。図3から明らかなように、焼結温度が $1100^\circ\text{C}$ の場合に $12.1\text{kG}$ の $Br$ 、 $25.4\text{kOe}$ の $iH_c$ 及び $35.1\text{MGoe}$ の $(BH)_{\text{max}}$ が得られ、焼結温度が $1110^\circ\text{C}$ の場合に $12.1\text{kG}$ の $Br$ 、 $25.2\text{kOe}$ の $iH_c$ 及び $35.0\text{MGoe}$ の $(BH)_{\text{max}}$ が得られ、 $Br$ 及び $(BH)_{\text{max}}$ は低かった。

【0074】この比較例の焼結磁石の組成を分析したところ、重量百分率で主成分は、Nd: 19.14%, Pr: 5.34%, Dy: 6.00%, B: 0.97%, Nb: 0.65%, Al: 0.10%

%, Co: 2.00%, Ga: 0.08%, Cu: 0.10%, 残部: Feであり、不純物はO: 0.17%, N: 0.06%, C: 0.06%であった。この比較例の焼結磁石の $Br$ 及び $(BH)_{\text{max}}$ が低いのは、Nb含有量が0.65%と高いので、主相結晶粒の焼結時の正常な粒成長が抑制されたためであると考えられる。

#### 【0075】実施例4

表8の主成分組成を有する溶製合金L及び溶製合金Mをそれぞれ用いた以外は実施例1と同様にして粗粉砕した。合金Lの粗粉 $90.0\text{kg}$ と合金Hの粗粉 $10.0\text{kg}$ とをV型混合機に投入して混合し、 $100\text{kg}$ の混合粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 22.83%, Pr: 6.37%, Dy: 1.30%, B: 1.05%, Mo: 0.13%, Al: 0.10%, 残部: Feであり、不純物はO: 0.15%, N: 0.01%, C: 0.02%であった。

#### 【0076】

【表8】

合金	組成(重量%)						
	Nd	Pr	Dy	B	Mo	Al	Fe
L	23.85	6.65	-	1.05	0.15	0.10	残部
M	13.68	3.82	13.00	1.05	-	0.10	残部

【0077】この混合粗粉を用いた以外は実施例1と同様にして、微粉砕（平均粒径 $4.0\mu\text{m}$ ）、スラリー化及び磁界中成形を行った。得られた成形体を約 $5 \times 10^{-1}$  Torrの真空中で $200^\circ\text{C}$ で1時間加熱して脱油し、引き続き約 $2 \times 10^{-5}$  Torrの真空中で $1050 \sim 1100^\circ\text{C}$ の温度範囲内

の各温度で2時間焼結後室温まで冷却した。得られた各焼結体に不活性ガス雰囲気中で $900^\circ\text{C} \times 2$ 時間と $550^\circ\text{C} \times 1$ 時間の熱処理を各1回施した後、室温まで冷却してR-T-B系焼結型永久磁石を得た。 $20^\circ\text{C}$ で磁気特性を測定した結果、焼結温度が $1060 \sim 1090^\circ\text{C}$ の場合に永久磁石とし



て好ましい磁気特性が得られた。特に焼結温度が1070℃の場合に13.9kGのBr、15.5kOeのiHc及び46.5MG0eの(BH)maxが得られ、焼結温度が1080℃の場合に14.0kGのBr、15.3kOeのiHc及び47.2MG0eの(BH)maxが得られ、Br及び(BH)maxが高かった。

【0078】前記永久磁石のうち代表的な永久磁石の組成を分析したところ、重量百分率で主成分組成がNd: 2.283%, Pr: 6.37%, Dy: 1.30%, B: 1.05%, Mo: 0.13%, Al: 0.10%, 残部: Feであり、不純物はO: 0.18%, N: 0.06%, C: 0.08%であった。

【0079】焼結温度1070℃、1080℃の条件で作製した前記永久磁石の断面組織について、後述の実施例7と同様にして主相結晶粒( $R_2T_{14}B$ )内(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy)の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy)の濃度が結晶粒界相より高い第一の主相結晶粒と、重希土

類元素(Dy)の濃度が結晶粒界相より低い第二の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相とほぼ等しい第三の主相結晶粒とから構成されていることが分かった。

#### 【0080】実施例5

表9の主成分組成を有する溶製合金N及び溶製合金Oをそれぞれ用いた以外は実施例1と同様にして粗粉碎した。合金Nの粗粉80.0kgと合金Oの粗粉20.0kgとをV型混合機に投入して混合し、100kgの粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 26.2%, Dy: 5.8%, B: 0.95%, Nb: 0.20%, Al: 0.1%, Co: 2.5%, Cu: 0.15%, Ga: 0.15%, 残部: Feであり、不純物はO: 0.15%, N: 0.02%, C: 0.02%であった。

#### 【0081】

【表9】

合金	組成 (重量%)								
	Nd	Dy	B	Nb	Al	Co	Cu	Ga	Fe
N	29.0	3.0	0.95	—	0.10	2.50	0.15	0.15	残部
O	15.0	17.0	0.95	1.00	0.10	2.50	0.15	0.15	残部

【0082】混合粗粉を酸素濃度が500ppm以下(体積比)の窒素ガス雰囲気中でジェットミル粉碎し、平均粒径 $4.2\mu m$ の微粉とした。この微粉を磁界強度10kOe、成形圧1.5ton/cm<sup>2</sup>の条件で乾式圧縮成形した。得られた成形体を約 $3 \times 10^{-5}$  Torrの真空中で、1040~1100℃の温度範囲内の各温度で2時間焼結後室温まで冷却した。

【0083】得られた各焼結体に不活性ガス雰囲気中で900℃×3時間と480℃×1時間の熱処理を各1回施した後、室温まで冷却してR-T-B系統結型永久磁石を得た。20℃で磁気特性を測定したところ、焼結温度を1050~1090℃とした場合に永久磁石として好ましい磁気特性が得られた。特に焼結温度が1070℃の時に12.5kGのBr、24.5kOeのiHc及び37.5MG0eの(BH)maxが得られ、焼結温度が1080℃の時に12.5kGのBr、24.2kOeのiHc及び37.4MG0eの(BH)maxが得られ、Br及び(BH)maxが高かった。前記永久磁石を分析したところ、重量百分率で主成分は、Nd: 26.2%, Dy: 5.8%, B: 0.95%, Nb: 0.20%, Al: 0.1%, Co: 2.5%, Cu: 0.15%, Ga: 0.15%, 残部: Feであり、不純物はO: 0.38%, N: 0.03%, C: 0.05%であった。

【0084】焼結温度が1070℃、1080℃の前記焼結磁石の断面組織について、後述の実施例7と同様にして主相結晶粒( $R_2T_{14}B$ )内(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy)の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy)の濃度が結晶粒界相より高い第一の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相より低い第二の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相とほぼ等しい第三の主相結晶粒とから構成されていることが分かった。

#### 【0085】実施例6

表10の主成分組成を有する溶製合金P及び溶製合金Qをそれぞれ用いた以外は実施例1と同様にして粗粉碎した。合金Pの粗粉90.0kgと合金Qの粗粉10.0kgとをV型混合機に投入して混合し、100kgの混合粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 20.6%, Pr: 8.8%, Dy: 2.6%, B: 1.06%, W: 0.18%, Al: 0.05%, Ga: 0.17%, 残部: Feであり、不純物はO: 0.15%, N: 0.01%, C: 0.01%であった。

#### 【0086】

【表10】

合金	組成 (重量%)							
	Nd	Pr	Dy	B	W	Al	Ga	Fe
P	21.70	9.30	1.00	1.08	0.20	0.05	0.17	残部
Q	10.50	4.50	17.00	1.06	—	0.05	0.17	残部

【0087】混合粗粉を酸素濃度が500ppm以下(体積比)の窒素ガス雰囲気中でジェットミル粉碎し、平均粒径 $4.2\mu m$ の微粉とした。この微粉を磁界強度10kOe、成形圧1.5ton/cm<sup>2</sup>の条件で乾式圧縮成形した。得られた成

形体を約 $3 \times 10^{-5}$  Torrの真空中で、1040~1100℃の温度範囲内の各温度で2時間焼結後、室温まで冷却した。

【0088】得られた各焼結体に不活性ガス雰囲気中で900℃×3時間と550℃×1時間の熱処理を各1回施した

後室温まで冷却し、R-T-B系統結晶型永久磁石を得た。20℃で磁気特性を測定したところ、焼結温度を1050～1090℃とした場合に永久磁石として好ましい磁気特性が得られた。特に焼結温度が1070℃のときに13.2kGのBr、19.5kOeのiHc及び41.8MG0eの(BH)maxが得られ、焼結温度が1080℃のときに13.2kGのBr、19.3kOeのiHc及び41.7MG0eの(BH)maxが得られ、Br及び(BH)maxが高かった。

【0089】前記永久磁石のうち代表的な永久磁石の組成を分析したところ、重量百分率でNd: 20.6%, Pr: 8.8%, Dy: 2.6%, B: 1.06%, W: 0.18%, Al: 0.05%, Ga: 0.17%, 残部: Feであり、不純物はO: 0.50%, N: 0.02%, C: 0.06%であった。

【0090】焼結温度1070℃及び1080℃の条件で作製した前記永久磁石の断面組織について、後述の実施例7と同様にして主相結晶粒( $R_2T_{14}B$ )内(ほぼ中心部)及び結晶粒界相における重希土類元素(Dy)の濃度を測定した。その結果、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy)

y)の濃度が結晶粒界相より高い第一の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相より低い第二の主相結晶粒と、重希土類元素(Dy)の濃度が結晶粒界相とほぼ等しい第三の主相結晶粒とから構成されていることが分かった。

#### 【0091】実施例7

表11の主成分組成を有する溶製合金R及び溶製合金Sを各々用いた以外は実施例1と同様にして粗粉碎した。合金Rの粗粉90.0kgと合金Sの粗粉10.0kgとをV型混合機に投入して混合し、100kgの混合粗粉とした。混合粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 21.38%, Pr: 7.12%, Dy: 1.50%, B: 1.03%, Al: 0.08%, Co: 2.00%, Ga: 0.08%, Cu: 0.1%, 残部: Feであり、O: 0.14%, N: 0.02%, C: 0.02%であった。

#### 【0092】

【表11】

合金	組成(%)								
	Nd	Pr	Dy	B	Al	Co	Ga	Cu	Fe
R	22.50	7.50	-	1.03	0.08	2.00	0.08	0.10	残部
S	11.25	3.75	15.00	1.03	0.08	2.00	0.08	0.10	残部

【0093】混合粗粉を酸素濃度が10ppm以下(体積比)の窒素ガス雰囲気中でジェットミル粉碎し、平均粒径 $4.2\mu m$ の微粉とした。得られた微粉を窒素ガス雰囲気中で、大気に触れさせずに直接鋤油(出光興産(株)製、商品名: 出光スーパーゾルPA-30)中に回収した。得られたスラリーを磁界強度10kOe、成形圧 $1.0\text{ton/cm}^2$ で圧縮成形した。得られた成形体を約 $5 \times 10^{-1}$  Torrの真空中で200℃で1時間加熱して脱油し、約 $3 \times 10^{-5}$  Torrの真空中で1040～1100℃の温度範囲内の各温度で2時間焼結後、室温まで冷却した。

【0094】各焼結体に不活性ガス雰囲気中で900℃×2時間と480℃×1時間の熱処理を各1回施した後、室温まで冷却してR-T-B系統結晶型永久磁石を作製した。20℃で磁気特性を測定したところ、焼結温度が1060～1090℃の場合に永久磁石として好ましい磁気特性が得られた。特に焼結温度が1070℃の場合に13.9kGのBr、15kOeのiHc及び46.5MG0eの(BH)maxが得られ、焼結温度が1080℃の場合に14.0kGのBr、14.8kOeのiHc及び47.2MG0eの(BH)maxが得られ、Br及び(BH)maxが高かった。

【0095】前記焼結磁石のうち代表的な焼結磁石を分析したところ、重量百分率で主成分は、Nd: 21.38%, Pr: 7.12%, Dy: 1.50%, B: 1.03%, Al: 0.08%, Co: 2.00%, Ga: 0.08%, Cu: 0.1%, 残部: Feであり、不純物はO: 0.16%, N: 0.06%, C: 0.06%であった。

【0096】前記焼結磁石のうち代表的な焼結磁石の断面組織を、EPMA(Electron Probe Micro-Analyzer; JEDL製、型式JXA-8800)を用いて、下記の条件で分析した。

加速電圧: 15kV、

試料吸収電流: 約 $4 \times 10^{-8}$  A、

1分析点当たりのX線取り込み時間(計数時間): 10msec、

分析点数: 縦(X)方向及び横(Y)方向がともに400点、

X及びY方向の各分析点の間隔:  $0.12\mu m$ 、

面分析のエリア:  $0.12\mu m \times 400\text{点} = 48\mu m$ の矩形範囲、

【0097】上記条件でビームを最小スポットに絞り照射することにより、Dy、Nd及びPrの濃度分布を測定した。Dy、Nd及びPrの分析に用いた分光結晶は高感度型ふっ化リチウム(LiF)であった。本実施例のR-T-B系統結晶型永久磁石の結晶組織を図4に概略的に示す。結晶組織は $R_2T_{14}B$ 型主相結晶粒1と結晶粒界相2とを有し、結晶粒界相の3重点2'は黒い領域で示す。また図4の結晶組織におけるDyの濃度分布を図5に示し、Ndの濃度分布を図6(a)に示し、Prの濃度分布を図6(b)に示す。図5、図6(a)、(b)から明らかなように、結晶粒界相ではNd、Dy、Prの分布は実質的に3重点で認められたが、これは3重点のみにNd、Dy、Prが分布しているわけではなく、3重点以外では結晶粒界相が非常に薄いので、Nd、Dy、Prの分布量が非常に少ないからである。

【0098】図4において、結晶粒界相の3重点を形成しているのはR(Nd、Dy、Pr)リッチ相である。図6(a)及び(b)からNd及びPrはほぼ同位置に存在することが分かる。また図5、図6(a)、(b)から、DyはNd及びPrとほぼ同じ結晶粒界相の領域にも存在するが、結晶粒界から

1.0 $\mu$ m以上離れた $R_2T_{14}B$ 型主相結晶粒内の部分(芯部)でも高濃度に存在することが分かった。

【0099】これらの観察結果から、結晶粒界相から主相結晶粒内の中心部に至るDy濃度分布に関し、3種類のDy濃度分布のパターンがあることが分かった。第一のパターンでは、結晶粒界相より主相結晶粒内の芯部の方がDy濃度が高い。第二のパターンでは、結晶粒界相のDy濃度が高く主相結晶粒内の芯部のDy濃度が低い。第三のパターンでは、結晶粒界相から主相結晶粒の中心部に至るDy濃度分布がほぼ均一である。図5において、結晶粒界相より主相結晶粒内の芯部の方がDy濃度が高い第一の主相結晶粒は6個であり、結晶粒界相よりDy濃度が低い第二の主相結晶粒は15個であり、結晶粒界相とほぼ等しいDy濃度を有する第三の主相結晶粒は19個であった。なお図5、図6(a)、(b)においてDy、Nd及びPrの濃度分布を評価する際に、顕微鏡観察用試料の作成時に形成されたボイドの影響を考慮した。また図4、図5及び図6(a)、(b)は断面組織の一例にすぎず、Dyの濃度分布を決定するには多数の視野における断面組織から求めたデータを平均する必要がある。このように本発明のR-T-B系統結晶型永久磁石は、主相結晶粒及び結晶粒界相において特徴的なDy濃度分布を有する。

【0100】前記永久磁石のうち代表的なものの主相結晶粒の粒径分布を図7に示す。図7の横軸は主相結晶粒の粒径範囲を示し、例えば「9~10 $\mu$ m」は主相結晶粒の粒径範囲が「9 $\mu$ m以上10 $\mu$ m未満」であることを意味する。主相結晶粒の粒径は、光学顕微鏡(型式UFX-I I、株式会社ニコン製)を用いて、永久磁石の任意の断面の写真(倍率1000倍)を撮影し、この断面写真をプラネトン社製の画像処理ソフト(Image Pro. Plus (DOS

V))により画像処理した。画像処理で測定した任意の各主相結晶粒の面積を $S_i$ として、さらに各主相結晶粒の断面形状を円と仮定し、各主相結晶粒径 $d_i$ を $(4 \times S_i \div \pi)^{1/2}$ と定義した。縦軸の分布率(%)は、測定した視野における主相結晶粒の総数 $T$ に対する各粒径範囲内にある主相結晶粒の個数 $T_N$ の比率 $[(T_N/T) \times 100\%]$ を示す。

【0101】図7から明らかなように、本発明の永久磁石では、粒径2 $\mu$ m未満の主相結晶粒の分布率が0%であり、かつ16 $\mu$ m以上の主相結晶粒の分布率が5.8%であった。さらに検討した結果、粒径2 $\mu$ m未満の主相結晶粒の分布率が5%未満でかつ16 $\mu$ m以上の主相結晶粒の分布率が10%以下であれば永久磁石として好ましい磁気特性を実現できることが分かった。さらに粒径2 $\mu$ m未満の主相結晶粒の分布率が3%以下でかつ16 $\mu$ m以上の主相結晶粒の分布率が8%以下であるのがより好ましく、粒径2 $\mu$ m未満の主相結晶粒の分布率が0%でかつ16 $\mu$ m以上の主相結晶粒の分布率が6%以下であるのが特に好ましいことが分かった。なお前記主相粒径分布はNb含有量が0.01~0.6%の場合でも実現可能である。

#### 【0102】比較例5

表12の主成分組成を有する溶製合金Tを用いた以外は実施例7と同様にして粗粉碎した。粗粉の組成を分析したところ、重量百分率で主成分は、Nd: 21.38%, Pr: 7.12%, Dy: 1.50%, B: 1.03%, Nb: 0.70%, Al: 0.08%, Co: 2.00%, Ga: 0.08%, Cu: 0.1%, 残部: Feであり、不純物はO: 0.15%, N: 0.01%, C: 0.02%であった。

#### 【0103】

#### 【表12】

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
T	21.38	7.12	1.50	1.03	0.70	0.08	2.00	0.08	0.10	残部

【0104】この粗粉を用いて実施例7と同様にして、微粉碎(平均粒径4.1 $\mu$ m)、スラリー化、磁界中成形、脱油、焼結及び熱処理を行い、シングル法による比較例の焼結型永久磁石を得た。この焼結磁石の組成を分析したところ、重量百分率で主成分は、Nd: 21.38%, Pr: 7.12%, Dy: 1.50%, B: 1.03%, Nb: 0.70%, Al: 0.08%, Co: 2.00%, Ga: 0.08%, Cu: 0.1%, 残部: Feであり、不純物はO: 0.17%, N: 0.05%, C: 0.07%であった。

【0105】20℃において磁気特性を測定した結果、iHcのレベルは16kOe前後と高いが、Brは13.5kG以下、(BH)max44.0MGoe以下であり、実施例7に比べて低かった。

【0106】図8はこの焼結磁石の断面組織を概略的に示す。結晶組織中の3はボイドを示し、その他の番号は図4と同じである。図8より、結晶粒界相から主相結晶粒の中心部まではほぼ均一なDy濃度分布と、結晶粒界相の

Dy濃度が高く主相結晶粒内のほぼ中心部のDy濃度が低い分布の2パターンが存在することが確認された。結晶粒界相とほぼ同じDy濃度分布を有する主相結晶粒は31個であり、結晶粒界相よりDy濃度が低い主相結晶粒は15個であった。しかし結晶粒界相より主相結晶粒内のほぼ中心部のDy濃度が高い分布は観察されなかった。

【0107】この比較例の焼結磁石の主相結晶粒径分布を実施例7と同様にして評価した結果を図9に示す。図9から明らかなように、この焼結磁石では粒径1 $\mu$ m以上2 $\mu$ m未満の主相結晶粒の分布率が12.5%であり、かつ図7の分布に比べて全体的に小粒径側に主相結晶粒径分布が大きくシフトしていて、主相結晶粒が十分粒成長していない。このため実施例7に比べてBr及び(BH)maxが低いと判断される。

【0108】上記実施例では重希土類元素がDyの場合を記載したが、Tb又はHoの場合も、Dyの場合とほぼ同様

に、芯部において結晶粒界相よりTb又はHoの濃度が高い主相結晶粒を有し、上記実施例と同様に高いBr及び(BH)maxを有するR-T-B系統結型永久磁石を得ることができる。

【0109】上記実施例では、同じR含有量でかつR元素を構成するDy、Nd等の比率のみが異なる以外は他の主成分が一致する2種のR-T-B系合金粉末、又は同じR含有量でかつR元素を構成するDy、Nd等の比率及びFeの一部を高融点金属元素(Nb等)で置換した以外は他の主成分が一致する2種のR-T-B系合金粉末を用いて混合することにより、特徴あるDy濃度分布を有する主相結晶粒を有し、かつ高いBr及び(BH)maxの用途に好適な主相結晶粒径分布を有するR-T-B系統結型永久磁石を安定に得ることができた。本発明では前記R-T-B系合金粉末として、3種以上のR-T-B系合金粉末を用いてもよい。またこれらR-T-B系合金粉末の混合は微粉段階で行ってもよい。

【0110】上記実施例のR-T-B系統結型永久磁石に各種の表面処理(Niめっき及び/又は電着エポキシ樹脂コーティング等)を施せば、各種の用途(ボイスコイルモータ又はCDピックアップ等のアクチュエータ、あるいは回転機等)に好適に用いることができる。

【0111】

【発明の効果】以上詳述したとおり、本発明のR-T-B系統結型永久磁石は、 $R_2T_{14}B$ 型主相結晶粒が、重希土類元素(Dy等)濃度が結晶粒界相より高い第一の $R_2T_{14}B$ 型主相結晶粒と、重希土類元素(Dy等)濃度が結晶粒界相より低い第二の $R_2T_{14}B$ 型主相結晶粒と、重希土類元素(Dy等)濃度が結晶粒界相とほぼ等しい第三の $R_2T_{14}B$ 型主相結晶粒とから構成されているので、シングル法により得られたR-T-B系統結型永久磁石と同程度に高いiHcを有するとともに、より高いBr及び(BH)maxを有する。したがって本発明のR-T-B系統結型永久磁石は、高いBr及び高

い(BH)maxが要求される用途に好適に用いることができる。

【図面の簡単な説明】

【図1】実施例1及び比較例1のR-T-B系統結型永久磁石について、焼結温度と磁気特性(Br, iHc)との関係を示すグラフである。

【図2】実施例2及び比較例2のR-T-B系統結型永久磁石について、焼結温度と磁気特性(Br, iHc)との関係を示すグラフである。

【図3】実施例3及び比較例3, 4のR-T-B系統結型永久磁石について、焼結温度と磁気特性(Br, iHc)との関係を示すグラフである。

【図4】実施例7のR-T-B系統結型永久磁石の結晶組織を示す概略図である。

【図5】実施例7のR-T-B系統結型永久磁石の結晶組織におけるDyの濃度分布を示すEPMA写真である。

【図6】実施例7のR-T-B系統結型永久磁石の結晶組織における重希土類元素の濃度分布を示し、(a)は結晶組織におけるNdの濃度分布を示すEPMA写真であり、(d)は結晶組織におけるPrの濃度分布を示すEPMA写真である。

【図7】実施例7のR-T-B系統結型永久磁石における主相結晶粒の粒径分布を示すグラフである。

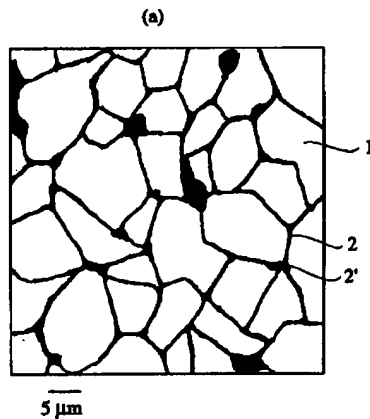
【図8】比較例5のR-T-B系統結型永久磁石の結晶組織を示す概略図である。

【図9】比較例5のR-T-B系統結型永久磁石における主相結晶粒の粒径分布を示すグラフである。

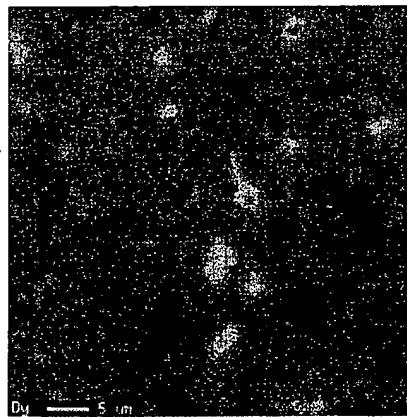
【符号の説明】

- 1・・・ $R_2T_{14}B$ 型主相結晶粒
- 2・・・結晶粒界相
- 2'・・・三重点
- 3・・・ボイド

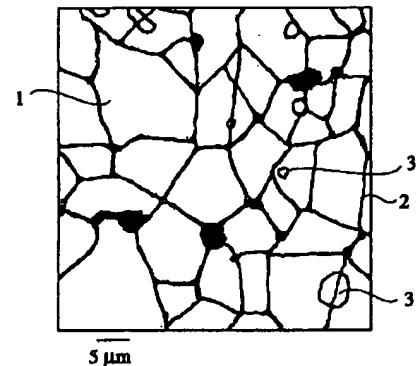
【図4】



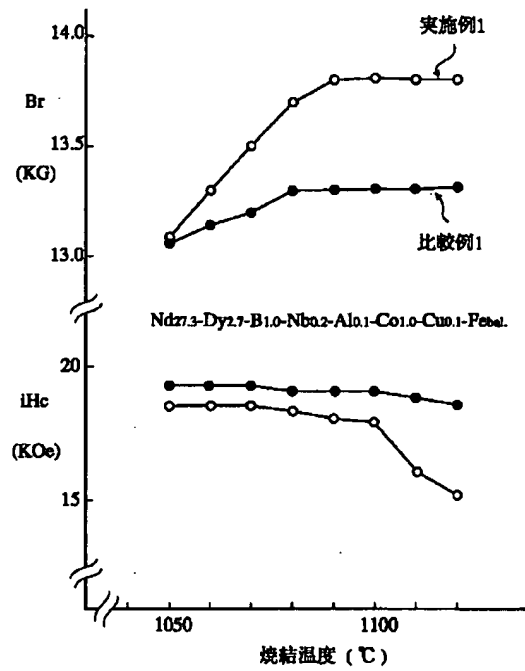
【図5】



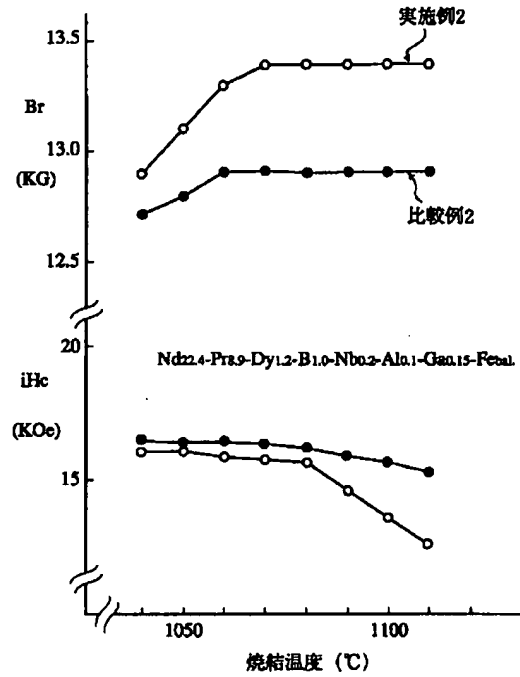
【図8】



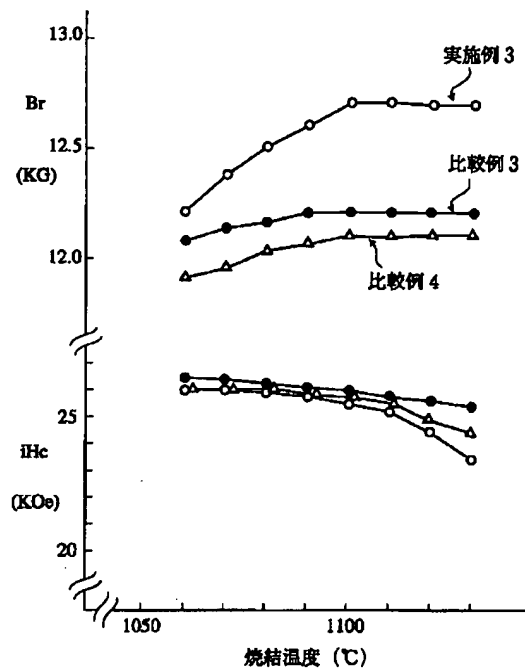
【図1】



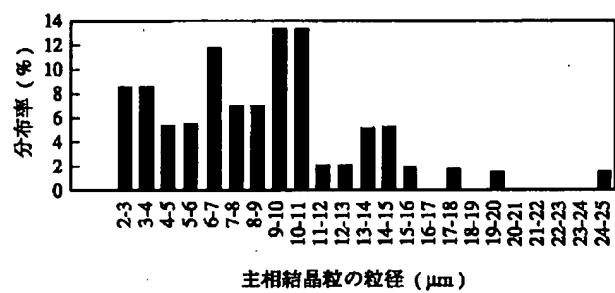
【図2】



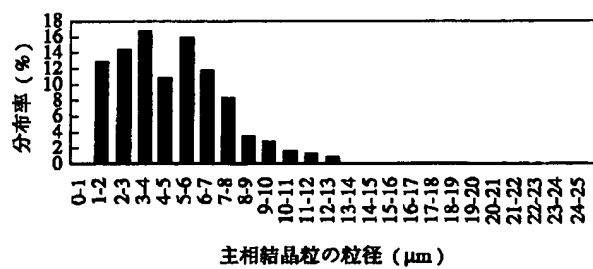
【図3】



【図7】

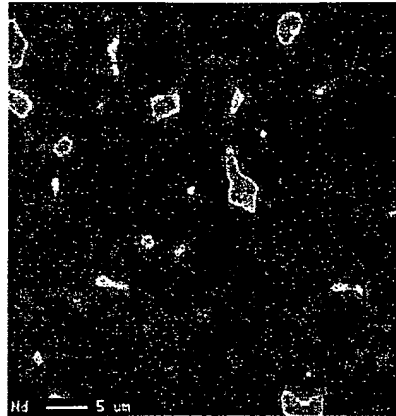


【図9】



【図6】

(a)



(b)



# PATENT ABSTRACTS OF JAPAN

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(22)Date of filing : 14.10.1999 (72)Inventor : UCHIDA KIMIO  
KAWADA TSUNEHIRO

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(54) R-T-B SINTERED PERMANENT MAGNET

(57)Abstract:

PROBLEM TO BE SOLVED: To provide an R-T-B sintered type permanent magnet of high performance which is suitable for a use requiring high Br and high (BH)max.

SOLUTION: This R-T-B sintered type permanent magnet (R is at least one kind

of rare earth element containing Y and necessarily contains at least one kind of heavy rare earth element selected from among a group composed of Dy, Tb and Ho, and T is Fe or Fe and Co) has a composition, where R: 28-33 wt.%, B: 0.5-2 wt.% and residue is practically composed of T and unavoidable impurities. The R-T-B sintered type permanent magnet has a texture containing first R2T14B type main phase crystal grains, where the concentration of heavy rare earth element is higher than these of crystal grain phase and second R2T14B type main phase crystal grains, where the concentration of the heavy rare earth element is lower than that in the crystal grain phase.

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#### LEGAL STATUS

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registration]

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[Date of registration]

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examiner's decision of rejection]

[Date of extinction of right]



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**CLAIMS**

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[Claim(s)]

[Claim 1] R-T-B system sintering mold permanent magnet which has the presentation which becomes an R0.5-2% and remainder real target from T and an unescapable impurity with weight percent: 28 - 33%, B: (R is a kind of rare earth elements containing Y at least) T is Fe, or Fe and Co, surely including at least a kind of heavy-rare-earth element chosen from the group which consists of Dy, Tb, and Ho. it is -- the R-T-B system sintering mold permanent magnet characterized by having an organization containing the first R2T14B type main phase crystal grain with the concentration of said heavy-rare-earth element higher than a grain boundary phase, and the second R2T14B type main phase crystal grain with the concentration of said heavy-rare-earth element lower than a grain boundary phase.

[Claim 2] The R-T-B system sintering mold permanent magnet characterized by containing 0.01 - 0.6% of M1 (however, M1 being a kind of element chosen from the group which consists of Nb, Mo, W, V, Ta, Cr, Ti, Zr, and Hf at least.) with weight percent in a R-T-B system sintering mold permanent magnet according to claim 1.

[Claim 3] The R-T-B system sintering mold permanent magnet characterized by containing 0.01 - 0.3% of M2 (however, M2 being a kind of element chosen from the group which consists of aluminum, Ga, and Cu at least.) with weight percent

in a R-T-B system sintering mold permanent magnet according to claim 1 or 2.

[Claim 4] The R-T-B system sintering mold permanent magnet with which R is characterized by being 33% or less exceeding 31% in a R-T-B system sintering mold permanent magnet according to claim 1 to 3 with weight percent.

[Claim 5] The R-T-B system sintering mold permanent magnet characterized by containing 0.6% or less of oxygen, 0.15% or less of carbon, 0.15% or less of nitrogen, and 0.3% or less of calcium with weight percent as said unescapable impurity in a R-T-B system sintering mold permanent magnet according to claim 1 to 4.

[Claim 6] The R-T-B system sintering mold permanent magnet characterized by containing 0.25% or less of oxygen, 0.15% or less of carbon, 0.03% or less of nitrogen, and 0.3% or less of calcium with weight percent as said unescapable impurity in a R-T-B system sintering mold permanent magnet according to claim 1 to 4.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the R-T-B system sintering mold permanent magnet which has high coercive force, a residual magnetic flux density, and a maximum energy product.

[0002]

[Description of the Prior Art] As for the R-T-B system sintering mold permanent magnet (R is a kind of rare earth elements containing Y at least, and T is Fe, or Fe and Co.), the thing of about 40 MGOe(s) is mass-produced by the maximum energy product. There are a single method and the blending method as a means to adjust the alloy presentation of a R-T-B system sintering mold permanent magnet.

[0003] The single method is an approach of performing grinding, shaping among a field, sintering, and heat treatment, and manufacturing a R-T-B system sintering mold permanent magnet using the ingot adjusted to the principal component presentation of a R-T-B system sintering mold permanent magnet in dissolution/casting phase, desired machining and surface treatment are given and practical use is presented with the obtained R-T-B system sintering mold permanent magnet.

[0004] The blending method is an approach of grinding if needed, performing shaping among a field, sintering, heat treatment, and surface treatment henceforth, and manufacturing a R-T-B system sintering mold permanent magnet, after mixing two or more sorts of alloy powder for R-T-B system sintering mold permanent magnets with which presentations differ with the compounding ratio which finally becomes the principal component presentation of a desired R-T-B system sintering mold permanent magnet.

[0005] According to the describing [ above ] single method, it is comparatively easy to heighten coercive force  $iH_c$ , but a residual magnetic flux density  $B_r$  and maximum energy product  $(BH)_{max}$  become low, and there is a problem of not being suitable in the application as which high  $B_r$  and high  $(BH)_{max}$  are required.

[0006] Moreover, although the R-T-B system sintering mold permanent magnet (JP,7-122413,A) which comes to blend a R-T system alloy with high R content

and a R-T-B system alloy with low R content as an example of application of the conventional blending method, and the R-T-B system sintering mold permanent magnet (JP,9-232121,A) which Ga, C, and O are segregating on R rich phase and the outskirts of it are proposed, the room of amelioration is to make it suitable for the application of high Br and high (BH) max. Nothing is solved about the optimal concentration distribution of the heavy-rare-earth element of the main phase crystal grain especially with the large effect on magnetic properties, and its control approach.

[0007] Therefore, the purpose of this invention is offering the R-T-B system sintering mold permanent magnet of the suitable high performance for the application as which high Br and high (BH) max are required.

[0008]

[Means for Solving the Problem] An example is taken by the above-mentioned purpose. Wholeheartedly as a result of research this invention persons The presentation which becomes a remainder real target from T and an unescapable impurity R 28-33% and B0.5-2% with weight percent (R is a kind of rare earth elements containing Y at least) T is Fe, or Fe and Co, surely including at least a kind of heavy-rare-earth element chosen from the group which consists of Dy, Tb, and Ho. It has. The first R2T14B type main phase crystal grain with the concentration of said heavy-rare-earth element higher than a grain boundary phase, The R-T-B system sintering mold permanent magnet which has an organization containing the second R2T14B type main phase crystal grain with the concentration of said heavy-rare-earth element lower than a grain boundary phase hit on an idea of high Br and high (BH) max being shown to header this invention.

[0009] the R-T-B system sintering mold permanent magnet by one desirable example of this invention -- weight percent -- R:28 - 33%, and B: 0.5 - 2%, and M1 : 0.01 - 0.6% (M1 is a kind of element chosen from the group which consists of Nb, Mo, W, V, Ta, Cr, Ti, Zr, and Hf at least.), and the remainder -- it has the presentation which consists of T and an unescapable impurity substantially.

[0010] The R-T-B system sintering mold permanent magnet by another desirable example of this invention It is R0.01-0.6% (M1 is a kind of element chosen from the group which consists of Nb, Mo, W, V, Ta, Cr, Ti, Zr, and Hf at least.) at weight percent. : 28 - 33%, B: 0.5 - 2%, M1 : M20.01-0.3% (M2 is a kind of element chosen from the group which consists of aluminum, Ga, and Cu at least.) : It has the presentation which becomes a remainder real target from T and an unescapable impurity.

[0011] R is 33% or less exceeding 31% in weight percent, and the R-T-B system sintering mold permanent magnet by the still more desirable example of this invention has the presentation which contains 0.6% or less of oxygen, 0.15% or less of carbon, 0.03% or less of nitrogen, and 0.3% or less of calcium as an unescapable impurity.

[0012] R is 28 - 31% in weight percent, and the R-T-B system sintering mold permanent magnet by the still more desirable example of this invention has the presentation which contains 0.25% or less of oxygen, 0.15% or less of carbon, 0.15% or less of nitrogen, and 0.3% or less of calcium as an unescapable impurity.

[0013] The R-T-B system sintering mold permanent magnet of this invention of the whole quantity of rare earth elements is the same, except that the ratios of heavy-rare-earth elements (Dy etc.) / light-rare-earth elements (Nd, Pr, etc.) differ, a presentation mixes two or more kinds of same alloy powder substantially, and it performs shaping among a field, sintering, and heat treatment, and it is obtained from subsequently performing machining, finish-machining (barrel finishing etc.), and surface treatment (nickel plating etc.) if needed. It is important to choose and have the optimal sintering conditions according to the presentation of two or more kinds of said alloy powder and the last presentation of a R-T-B system sintering mold permanent magnet, and to control strictly the diffusion condition of the heavy-rare-earth elements (Dy etc.) in a sintered compact in-house. Consequently, the crystalline structure containing the R2T14B type main phase crystal grain with the concentration of heavy-rare-earth elements (Dy etc.) higher

than a grain boundary phase and the R2T14B type main phase crystal grain with the concentration of heavy-rare-earth elements (Dy etc.) lower than a grain boundary phase is obtained about concentration distribution of the R2T14B type main phase crystal grain (almost core) and the heavy-rare-earth elements (Dy etc.) in a grain boundary phase.

[0014] although coercive force  $iH_c$  is a little low compared with the R-T-B system sintering mold permanent magnet by the single method, the R-T-B system sintering mold permanent magnet which has such a sintered compact organization is boiled markedly, and has high  $B_r$  and high  $(BH)_{max}$ . Although the correlation with this and concentration distribution of heavy-rare-earth elements (Dy etc.) is not clear still enough, the R2T14B type main phase crystal grain with heavy-rare-earth element concentration (Dy etc.) higher than a grain boundary phase contributes to high implementation of  $B_r$ , and is presumed to be what has contributed to the high implementation of  $iH_c$  with the R2T14B type main phase crystal grain near the single method with heavy-rare-earth element concentration (Dy etc.) lower than a grain boundary phase.

[0015]

[Embodiment of the Invention] [1] The presentation of the R-T-B system sintering mold permanent magnet of R-T-B system sintering mold (permanent magnet A) (presentation a) principal component this invention consists of the principal component and unescapable impurity which consist of R 28-33%, B 0.5-2%, and T with weight percent. In addition, it is desirable as a principal component to contain 0.01 - 0.6% of the weight of M1 (at least a kind of element chosen from the group which consists of Nb, Mo, W, V, Ta, Cr, Ti, Zr, and Hf), and/or 0.01 - 0.3% of the weight of M2 (at least a kind of element chosen from the group which consists of aluminum, Ga, and Cu).

[0016] (1) At least, R element R elements are a kind of rare earth elements containing Y, and surely contain a kind of heavy-rare-earth [ at least ] element chosen from the group which consists of Dy, Tb, and Ho. As rare earth elements other than a heavy-rare-earth element (Y is included), Nd, Pr, La, Sm, Ce, Eu,

Gd, Er, Tm, Yb, Lu, and Y are mentioned. As rare earth elements R, the mixture of two or more sorts of rare earth elements may be used like a misch metal or didym.

[0017] The content of R is 28 - 33 % of the weight. The fall of Br will become remarkable, if high iHc which is equal to practical use in the content of R being less than 28 % of the weight is not obtained and it exceeds 33 % of the weight.

[0018] As for the content of a heavy-rare-earth element, it is desirable that it is 0.2 - 15% of the weight of within the limits. The improvement effectiveness of magnetic properties according that the content of a heavy-rare-earth element is less than 0.2 % of the weight to distribution of the heavy-rare-earth element in the crystalline structure is inadequate. Moreover, if the content of a heavy-rare-earth element exceeds 15 % of the weight, Br of a R-T-B system sintering mold permanent magnet and (BH) max will fall greatly. The content of a more desirable heavy-rare-earth element is 0.5 - 13 % of the weight.

[0019] (2) The content of BB is 0.5 - 2 % of the weight. The fall of Br will become remarkable, if it is difficult to obtain high iHc which is equal to practical use in the content of B being less than 0.5 % of the weight and it exceeds 2 % of the weight.

[0020] (3) A T element T element is a Fe independent or Fe+Co. While the corrosion resistance of a sintering mold permanent magnet is improved by addition of Co, the Curie point goes up and the thermal resistance as a permanent magnet improves. However, if the content of Co exceeds 5 % of the weight, a Fe-Co phase harmful to the magnetic properties of a R-T-B system sintering mold permanent magnet will be formed, and both Br and iHc will fall. Therefore, Co content is made into 5 or less % of the weight. On the other hand, less than 0.5 % of the weight of the improvement effect and the heat-resistant improvement effectiveness of corrosion resistance [ content / Co ] is insufficient. Therefore, when adding Co, it is desirable to make Co content into 0.5 - 5 % of the weight.

[0021] (4) M1 element M1 is at least a kind of refractory metal element chosen from the group which consists of Nb, Mo, W, V, Ta, Cr, Ti, Zr, and Hf. By

existence of M1 element, too much growth of the main phase crystal grain produced by diffusion of heavy-rare-earth elements (Dy etc.) in a sintering process is suppressed, it is stabilized and high iHc near the single method can be obtained. However, if M1 element is added superfluously, normal grain growth of the main phase crystal grain will be checked conversely, and the fall of Br will be caused. Therefore, the upper limit of the content of M1 element is 0.6 % of the weight. On the other hand, addition effectiveness with the content of M1 element sufficient at less than 0.01 % of the weight is not accepted. Therefore, as for the content of M1 element, it is desirable that it is 0.01 - 0.6 % of the weight.

[0022] (5) M2 element M2 is a kind of element chosen from the group which consists of aluminum, Ga, and Cu at least.

[0023] iHc of a R-T-B system sintering mold permanent magnet and corrosion resistance are improved by minute amount addition of aluminum. However, since Br will fall greatly if aluminum content exceeds 0.3 % of the weight, aluminum content is made into 0.3 or less % of the weight. On the other hand, less than 0.01 % of the weight of iHc or a corrosion resistance improvement effect is [ aluminum content ] insufficient.

[0024] iHc of a R-T-B system sintering mold permanent magnet improves notably by minute amount addition of Ga. However, since Br will fall sharply like aluminum if it exceeds 0.3 % of the weight, Ga content is made into 0.3 or less % of the weight. Moreover, the meaningful improvement effectiveness of iHc is not accepted at less than 0.01 % of the weight.

[0025] Minute amount addition of Cu is effective in the corrosion resistance improvement of a sintered compact, and improvement in iHc. However, like aluminum and Ga, if Cu content exceeds 0.3 % of the weight, Br of a R-T-B system sintering mold permanent magnet falls sharply, and less than 0.01% of a corrosion resistance improvement and the improvement effectiveness of iHc is insufficient.

[0026] The content of M2 element makes either aluminum Ga or Cu 0.01 - 0.3 % of the weight as above.



[0027] (b) Oxygen, carbon, nitrogen, calcium, etc. are mentioned as an unescapable impurity unescapable impurity. calcium is mixed as an unescapable impurity, when two or more kinds of R-T-B system alloys with which the contents of a heavy-rare-earth element differ are produced by the reduction diffusion method (how to return the oxide powder of rare earth elements with a reducing agent (calcium), and to obtain alloy powder by counter diffusion with other basis metals continuously).

[0028] As for the content of oxygen, it is desirable that it is 0.6 or less % of the weight, as for a carbonaceous content, it is desirable that it is 0.15 or less % of the weight, as for the content of nitrogen, it is desirable that it is 0.15 or less % of the weight, and, as for the content of calcium, it is desirable that it is 0.3 or less % of the weight. If the content of each unescapable impurity exceeds the above-mentioned upper limit, the magnetic properties of a R-T-B system sintering mold permanent magnet will fall. As a content of a more desirable unescapable impurity, oxygen is 0.25 or less % of the weight, carbon is 0.15 or less % of the weight, and nitrogen is 0.03 or less % of the weight. Oxygen is 0.05 - 0.25 % of the weight, carbon is 0.01 - 0.03%, and the nitrogen of especially the content of a desirable unescapable impurity is 0.02 - 0.15%.

[0029] The following are mentioned as an example of a presentation of a R-T-B system sintering mold permanent magnet of having such an amount of unescapable impurities.

[0030] (i) Presentation whose carbon R is 33% or less exceeding 31% in weight percent, oxygen is 0.6% or less, and is 0.15% or less, whose nitrogen is 0.03% or less and whose calcium is 0.3% or less. For example, carbon can be made and nitrogen can be made 0.005 - 0.03% for oxygen 0.01 to 0.15% 0.25 to 0.6% by adoption of a dry pressing method.

[0031] (ii) Presentation whose carbon R is 28 - 31% in weight percent, oxygen is 0.25% or less, and is 0.15% or less, whose nitrogen is 0.15% or less and whose calcium is 0.3% or less. For example, carbon can be made and nitrogen can be made 0.02 - 0.15% for oxygen 0.01 to 0.15% 0.05 to 0.25% of the weight by

adoption of a wet compaction method.

[0032] (B) The crystalline structure of the R-T-B system sintering mold permanent magnet of organization this invention has the R2T14B type main phase crystal grain and a grain boundary phase. Said R2T14B type main phase crystal grain is the first R2T14B type main phase crystal grain with the concentration of (i) heavy-rare-earth element higher than a grain boundary phase at least, The second R2T14B type main phase crystal grain with the concentration of (ii) heavy-rare-earth element lower than a grain boundary phase is contained. Said R2T14B type main phase crystal grain is . (iii) The concentration of a heavy-rare-earth element may contain the third main phase crystal grain almost equal to a grain boundary phase. In a core (core part), it measures mostly, and the concentration of the heavy-rare-earth element in the R2T14B type main phase crystal grain says the field of the R2T14B type main phase crystal grain which 1.0 micrometers or more of core parts of the R2T14B type main phase crystal grain entered from the grain boundary here. Although Dy is desirable as a heavy-rare-earth element, you may be the mixture of Tb and/or Ho, or them and Dy.

[0033] In the taken cross-section photograph of the crystalline structure, the total number of the R2T14B type main phase crystal grain is made into 100%. The ratio of the number of the first R2T14B type main phase crystal grain is 1 - 35%, the ratio of the number of the second R2T14B type main phase crystal grain is 3 - 55%, and, as for the ratio of the number of the third R2T14B type main phase crystal grain, it is desirable that it is 96 - 10%. first - the ratio of the number of the third R2T14B type main phase crystal grain -- the above -- it is difficult for a R-T-B system sintering mold permanent magnet to have the high coercive force  $iH_c$ , a residual magnetic flux density  $B_r$ , and maximum energy product  $(BH)_{max}$  as it is out of range. More preferably, the ratio of the number of the first R2T14B type main phase crystal grain is 3 - 30%, the ratio of the number of the second R2T14B type main phase crystal grain is 10 - 45%, and the ratio of the number of the third R2T14B type main phase crystal grain is 87 - 25%.

[0034] [2] In order to manufacture the R-T-B system sintering mold permanent magnet of this invention which has the manufacture approach above-mentioned organization, adopt the so-called blending method which mixes two or more kinds of R-T-B system alloy powder with which the contents of heavy-rare-earth elements, such as Dy, differ. in this case, every -- it is made for the total quantity of R element to become the same with each alloy powder as for the presentation of R-T-B system alloy powder For example, in Nd+Dy, as shown in the below-mentioned example 1, one alloy powder is set to 29.0%Nd+1.0%Dy, and the alloy powder of another side is set to 15.0%Nd+15.0%Dy. About elements other than R element, although it is desirable that each alloy powder is substantially the same, some difference may be in the content of M1 and/or M2.

[0035] for example, the content of the heavy-rare-earth [ when mixing two kinds of alloy powder, while making the total quantity of both R element the same, make the content of the heavy-rare-earth element in the first alloy powder into 0 - 10 % of the weight, and ] element in the second alloy powder -- 10 % of the weight -- super- -- it may be 40 or less % of the weight. In this case, it is desirable to set the compounding ratio of the first alloy powder / second alloy powder to 70 / 30 - 95/5 by weight, and it is more desirable to be referred to as 80 / 20 - 90/10. This, so that the difference of the content of the heavy-rare-earth element between the first alloy powder and the second alloy powder becomes large The difference in the pulverizing nature between the first alloy powder and the second alloy powder (particle size distribution of fines) becomes large. It is because the square shape nature of a demagnetization curve and degradation of (BH) max which show the relation of intensity-of-magnetization (4 $\pi$ il)-magnetic field strength (H) by the particle size distribution of the main phase crystal grain of the R-T-B system sintering mold permanent magnet finally obtained becoming broad are invited.

[0036] Pulverizing of R-T-B system alloy powder can be performed by wet-grinding methods, such as dry grinding methods, such as a jet mill through inert gas, or a ball mill. In order to acquire high magnetic properties, it is desirable to

collect fines out of an inert gas ambient atmosphere in direct mineral oil, synthetic oil, vegetable oil, or those mixed oils, as after jet mill pulverizing and atmospheric air cannot be touched in the inert gas (concentration: it is 1000 ppm at volume ratio following) ambient atmosphere which does not contain oxygen substantially, and to make it mixture (slurry). Oxidation and adsorption of moisture can be controlled by intercepting fines from atmospheric air. As mineral oil, synthetic oil, or vegetable oil, from a viewpoint of deoiling nature and a moldability, a thing 350 degrees C or less has a desirable point distilling fractionally, in a room temperature, the thing of kinematic viscosity of 10 or less cSts is good, and its thing of 5 or less cSts is more desirable.

[0037] As two or more sorts of alloys for R-T-B system sintering mold permanent magnets blended in order to manufacture the permanent magnet of this invention, the sheet metal-like alloy (strip cast alloy) illustrated by the Japan patent No. 2,665,590, the Japan patent No. 2,745,042, etc. may be used. This sheet metal-like alloy (strip cast alloy) quenches and comes to solidify the alloy molten metal which has the presentation with which the requirements for this invention are filled with molten metal quenching methods, such as the single rolling method, the congruence rolling method, or a rotation disc method, and it has the homogeneous organization of a columnar crystal mostly, and the diameter of average crystal grain of the direction of a minor axis of said columnar crystal is 3-20 micrometers. High Br and (BH) Grinding is desirable after performing homogenization heat treatment which cools a sheet metal-like alloy in inert gas ambient atmospheres (Ar etc.) to an after [ 900-1200 degree-Cx1 - 10 hour heating ] room temperature, in order to obtain max.

[0038] A Plastic solid is acquired using mixture (slurry) by carrying out a wet compaction in a field with desired shaping equipment. In order to suppress degradation of the magnetic properties by oxidation, it is desirable to hold in an oil or an inert gas ambient atmosphere in between [ until it puts into a sintering furnace from immediately after shaping ]. Shaping may be performed with dry process. In the case of a dry pressing method, press forming of the mixture of

desiccation fines is carried out all over a magnetic field in an inert gas ambient atmosphere.

[0039] If a temperature up is rapidly carried out from ordinary temperature to sintering temperature on the occasion of sintering of a wet compaction object, the mineral oil, synthetic oil, or vegetable oil which remained in the Plastic solid will react with rare earth elements, will generate rare earth carbide, and will cause degradation of the magnetic properties of the sintered magnet obtained. It is desirable to perform deoiling processing held 30 minutes or more by the temperature of 100-500 degrees C and below degree of vacuum 10<sup>-1</sup> Torr as this cure. The mineral oil, synthetic oil, or vegetable oil which remains in a Plastic solid by deoiling processing is fully removable. In addition, whenever [stoving temperature] does not need to be fixed if it is a 100-500-degree C temperature requirement. Moreover, while carrying out a temperature up from a room temperature to 500 degrees C with the degree of vacuum below 10<sup>-1</sup> Torr, the deoiling effectiveness almost equivalent also as the following can be hereafter acquired for a programming rate by 5-degree-C/preferably by 10-degree-C/.

[0040] By sintering a Plastic solid at the temperature of about 1000-1200 degrees C in an inert gas ambient atmosphere, a R-T-B system sintering mold permanent magnet is manufactured. Desired machining and surface treatment are performed to the obtained R-T-B system sintering mold permanent magnet. As surface preparation, nickel plating, electrodeposited epoxy resin coating, etc. are mentioned.

[0041]

[Example] Although the following examples explain this invention to a detail further, this invention is not limited to them.

[0042] Coarse powder with a particle size of 500 micrometers or less was obtained by carrying out coarse grinding of the ingot alloy A which has the principal component presentation of example 1 table 1, and the ingot alloy B, and carrying out screen analysis in an inert gas ambient atmosphere, respectively.

87.9kg of coarse powder of Alloy A and 12.1kg of coarse powder of Alloy B were

thrown into the V shaped rotary mixer, it mixed, and 100kg mixed coarse powder was obtained. When the presentation of mixed coarse powder was analyzed, the impurities which principal components are Nd27.3%, Dy2.7%, B1.0%, and Nb0.2%, aluminum0.1%, Co1.0%, Cu0.1%, and Remainder Fe, and are contained in this mixed coarse powder with weight percent were 0.15% of the weight of O and 0.01% of the weight of N, and 0.02% of the weight of C.

[0043]

[Table 1]

合金	組成 (重量%)							
	Nd	Dy	B	Nb	Al	Co	Cu	Fe
A	29.0	1.0	1.0	0.2	0.1	1.0	0.1	残部
B	15.0	15.0	1.0	0.2	0.1	1.0	0.1	残部

[0044] The oxygen density carried out jet mill grinding in nitrogen-gas-atmosphere mind 10 ppm or less (volume ratio), and used said mixed coarse powder as fines with a mean particle diameter of 4.0 micrometers. Fines were collected in the state of atmospheric air and non-contact in nitrogen-gas-atmosphere mind in direct mineral oil (the Idemitsu Kosan make, trade name:Idemitsu super sol PA-30), and the fines slurry was obtained. the Plastic solid acquired using this fines slurry by carrying out wet compression molding on condition that magnetic-field-strength 10kOe and moulding pressure 1.0 ton/cm2 -- about -- the inside of the vacuum of  $5 \times 10^{-1}$  Torr -- 200 degrees C -- 1 hour -- heating -- after deoiling -- succeedingly -- about -- it sintered by  $3 \times 10^{-5}$  Torr in the 1050-1100-degree C temperature requirement for each 2 hours, it cooled to the room temperature, and the sintered compact was obtained.

[0045] After performing heat treatment of 900 degree-Cx 2 hours, and 500 degree-Cx 1 hour to each sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was obtained. When magnetic properties were measured in 20 degrees C, the result shown in drawing 1 was obtained. When sintering temperature was made into 1070-1110 degrees C so that clearly from drawing 1 , magnetic properties desirable as a permanent magnet were acquired.

When (BH) max of iHc of Br of 13.8kG(s) and 18kOe and 45.9MGOe(s) is obtained when especially sintering temperature is made into 1090 degrees C, and sintering temperature is made into 1100 degrees C, they are Br of 13.8kG(s), iHc of 17.9kOe, and 45.7MGOe. (BH) max was obtained and Br and (BH) max were high.

[0046] When the presentation of a typical sintered magnet was analyzed among said sintered magnets, principal components were Nd0.2%, aluminum:0.1%, Co:1.0%, Cu:0.1%, and Remainder: Fe in weight percent, and unescapable impurities were 0.17% of O and 0.05% of N, and 0.07% of C. : 27.3%, Dy : 2.7%, B: 1.0%, Nb :

[0047] It gazed at the cross-section organization of a typical sintered magnet like the below-mentioned example 7 among said sintered magnets, and the concentration of the heavy-rare-earth element (Dy) in the inside (almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase was measured. Consequently, it turned out that it consists of the first main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, the second main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase, and the third main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase. [0048] Coarse grinding was carried out like the example 1 except having used the ingot alloy C which has the principal component presentation of example of comparison 1 table 2. When the presentation (% of the weight) of this coarse powder was analyzed, principal components were Nd:0.1%, Co:1.0%, Cu:0.1%, and Remainder: Fe, and impurities were O:0.13%, N:0.008%, and C:0.02%. : 27.3%, Dy : 2.7%, B: 1.0%, Nb : 0.2%, aluminum

[0049]

[Table 2]

合金	組成 (重量%)							
	Nd	Dy	B	Nb	Al	Co	Cu	Fe
C	27.3	2.7	1.0	0.2	0.1	1.0	0.1	殘部

[0050] Pulverizing (mean particle diameter of 4.1 micrometers), slurring, shaping among a field, deoiling, sintering, and heat treatment were performed like the example 1 using this coarse powder, and the sintering mold permanent magnet of the example of a comparison by the single method was obtained. When the presentation (% of the weight) of this sintering mold permanent magnet was analyzed, principal components were Nd0.2%, aluminum:0.1%, Co:1.0%, Cu:0.1%, and Remainder: Fe, and impurities were O:0.15%, N:0.04%, and C:0.06%. : 27.3%, Dy : 2.7%, B: 1.0%, Nb :

[0051] The result of having measured magnetic properties in 20 degrees C is shown in drawing 1 R> 1. Although the level of iHc is as higher as 19kOe(s) order than drawing 1 , Br is 13.3 or less kGs, (BH) max is 42.5 or less MGOes, and it turns out that it is low compared with Br of an example 1, and (BH) max. Moreover, the main phase crystal grain with the concentration of the heavy-rare-earth element Dy higher than a grain boundary phase was not observed by the cross-section organization of the sintered magnet of this example of a comparison. [0052] Coarse grinding of the ingot alloy D which has the principal component presentation of example 2 table 3, and the ingot alloy E was carried out like the example 1 except having used, respectively. 94kg of coarse powder of Alloy D and 6kg of coarse powder of Alloy E were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg mixed coarse powder. When the presentation of mixed coarse powder was analyzed, principal components were Nd:0.15% and Remainder: Fe in weight percent, and impurities were O:0.14%, N:0.01%, and C:0.01%. : 22.4%, Pr: 8.9%, Dy : 1.2%, B: 1.0%, aluminum: 0.1%, Ga

[0053]

[Table 3]



合金	組成 (重量%)						
	Nd	Pr	Dy	B	Al	Ga	Fe
D	23.2	9.3	—	1.0	0.1	0.15	残部
E	8.9	3.6	20.0	1.0	0.1	0.15	残部

[0054] The oxygen density carried out jet mill grinding in the nitrogen-gas-atmosphere mind below 500 ppm (volume ratio), and used mixed coarse powder as fines with a mean particle diameter of 4.1 micrometers. Dry type compression molding was carried out on condition that magnetic-field-strength 10kOe and moulding pressure 1.5 ton/cm<sup>2</sup> using these fines. After sintering the acquired Plastic solid respectively among the vacuum of abbreviation 3x10<sup>-5</sup> Torr in a 1040-1110-degree C temperature requirement for 2 hours, it cooled to the room temperature and the sintered compact was obtained.

[0055] After performing heat treatment of 900 degree-Cx 3 hours, and 550 degree-Cx 1 hour to each sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was obtained. When magnetic properties were measured at 20 degrees C, the result shown in drawing 2 was obtained. Drawing 2 R> 2 showed that magnetic properties desirable as a permanent magnet were acquired when sintering temperature is made into 1050-1100 degrees C. When especially sintering temperature was 1070 degrees C, (BH) max of Br of 13.4kG(s), iHc of 16.3kOe, and 43.2MGoe(s) was obtained, and when sintering temperature was 1080 degrees C, (BH) max of Br of 13.4kG(s), iHc of 15.1kOe, and 43.3MGoe(s) was obtained, and Br and (BH) max were high.

[0056] When the presentation of a typical sintered magnet was analyzed among said sintered magnets, principal components were Nd:0.15% and Remainder: Fe in weight percent, and impurities were O:0.45%, N:0.02%, and C:0.07%. : 22.4%, Pr: 8.9%, Dy : 1.2%, B: 1.0%, aluminum: 0.1%, Ga

[0057] The concentration of a heavy-rare-earth element [ in / for the cross-section organization of a typical sintered magnet / the inside (almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase ] (Dy) was measured like the below-mentioned example 7 among said sintered magnets.

Consequently, the first R2T14B type main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, The second R2T14B type main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase and the third R2T14B type main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase showed being constituted.

[0058] Coarse grinding was carried out like the example 1 except having used the ingot alloy F which has the principal component presentation of example of comparison 2 table 4. When the presentation of coarse powder was analyzed, principal components were Nd:0.15% and Remainder: Fe in weight percent, and impurities were O:0.14%, N:0.01%, and C:0.02%. : 22.4%, Pr: 8.9%, Dy : 1.2%, B: 1.0%, aluminum: 0.1%, Ga

[0059]

[Table 4]

合金	組成 (重量%)						
	Nd	Pr	Dy	B	Al	Ga	Fe
F	22.4	8.9	1.2	1.0	0.1	0.15	残部

[0060] Pulverizing (mean particle diameter of 4.0 micrometers), shaping among a field, sintering, and heat treatment were performed like the example 2 using this coarse powder, and the sintering mold permanent magnet of the example of a comparison by the single method was obtained. When the component of this magnet was analyzed, principal components were Nd:0.15% and Remainder: Fe in weight percent, and impurities were O:0.43%, N:0.03%, and C:0.06%. : 22.4%, Pr: 8.9%, Dy : 1.2%, B: 1.0%, aluminum: 0.1%, Ga

[0061] The result of having measured magnetic properties at 20 degrees C is shown in drawing 2 . Although the level of iHc was a little high compared with the example 2 so that clearly from drawing 2 , Br was 12.9 or less kGs and (BH) max was as low as 40.1 or less MGOes. Moreover, the main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) higher than a grain

boundary phase was not observed by the cross-section organization of the sintered magnet of this example of a comparison. [0062] Coarse grinding was carried out like the example 1 except having used the ingot alloy G which has the principal component presentation of example 3 table 5, and the ingot alloy H. Next, 81.8kg of coarse powder of Alloy G and 18.2kg of coarse powder of Alloy H were thrown into the V shaped rotary mixer, it mixed, and 100kg mixed coarse powder was obtained. Remainder: When the presentation of mixed coarse powder was analyzed, in weight percent, Nd6.00%, B:0.97%, Nb:0.29%, aluminum:0.10%, Co:2.00%, Ga:0.08%, and Cu:0.10%, the principal component was Fe and were impurity O:0.14%, N:0.01%, and C:0.02%. : 19.14%, Pr: 5.34%, Dy :

[0063]

[Table 5]

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
G	22.29	6.21	2.00	0.97	0.35	0.10	2.00	0.08	0.10	殘部
H	5.03	1.47	24.00	0.97	—	0.10	2.00	0.08	0.10	殘部

[0064] Pulverizing (mean particle diameter of 4.2 micrometers), slurring, and compression molding in a field were performed like the example 1 using this mixed coarse powder. the acquired Plastic solid -- about -- the inside of the vacuum of  $5 \times 10^{-1}$  Torr -- 200 degrees C -- 1 hour -- heating -- deoiling -- subsequently -- about -- it cooled to the room temperature after 2-hour sintering at each temperature in the vacuum of  $2 \times 10^{-5}$  Torr, and in a 1060-1130-degree C temperature requirement. After performing heat treatment of 900 degree-Cx 2 hours, and 500 degree-Cx 1 hour to each obtained sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was obtained. The result of having measured magnetic properties in 20 degrees C is shown in drawing 3 . When sintering temperature was made into 1070-1120 degrees C so that clearly from drawing 3 , magnetic properties desirable as a permanent magnet were acquired. When especially sintering temperature was made into 1100 degrees C, (BH) max

of iHc of Br of 12.7kG(s) and 25.5kOe and 38.8MGOe(s) was obtained, when it considered as 1110 degrees C, (BH) max of iHc of Br of 12.7kG(s) and 25.3kOe and 38.6MGOe(s) was obtained, and Br and (BH) max were high.

[0065] When the presentation of a typical permanent magnet was analyzed among said permanent magnets, principal components were Nd6.00%, B:0.97%, Nb:0.29%, aluminum:0.10%, Co:2.00%, Ga:0.08%, Cu:0.10%, and Remainder Fe in weight percent, and impurities were O:0.16%, N:0.05%, and C:0.07%. : 19.14%, Pr: 5.34%, Dy :

[0066] The concentration of the heavy-rare-earth element (Dy) in the inside (almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase was measured like [ organization / of said permanent magnet produced on the sintering temperature of 1100 degrees C, and 1110-degree C conditions / cross-section ] the below-mentioned example 7. Consequently, it turned out that it consists of the first main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, the second main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase, and the third main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase. [0067] Coarse powder was obtained like the example 1 except having used the ingot alloy I which has the principal component presentation of example of comparison 3 table 6. When the presentation of this coarse powder was analyzed, principal components were Nd:6.00%, B:0.97%, Nb:0.29%, aluminum:0.10%, Co:2.00%, Ga:0.08%, Cu:0.10%, and Remainder: Fe in weight percent, and impurities were O:0.12%, N:0.01%, and C:0.01%. : 19.14%, Pr: 5.34%, Dy

[0068]

[Table 6]

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
I	19.14	5.34	6.00	0.97	0.29	0.10	2.00	0.08	0.10	殘部

[0069] Pulverizing (mean particle diameter of 4.2 micrometers), slurring, and

shaping among a field were performed like the example 1 except having used this coarse powder. To the acquired Plastic solid, deoiling, sintering, and heat treatment were performed on an example 3 and these conditions, and the sintering mold permanent magnet of the example of a comparison by the single method was obtained. When the presentation of this magnet was analyzed, principal components were Nd:6.00%, B:0.97%, Nb:0.29%, aluminum:0.10%, Co:2.00%, Ga:0.08%, Cu:0.10%, and Remainder: Fe in weight percent, and impurities were O:0.14%, N:0.04%, and C:0.06%. : 19.14%, Pr: 5.34%, Dy [0070] The result of having measured magnetic properties at 20 degrees C is shown in drawing 3 . Although the level of iHc was as high as 25kOe(s) order so that clearly from drawing 3 , Br was 12.2 or less kGs, (BH) max was 35.7 or less MGOes, and it was low compared with the example 3. Moreover, in the cross-section organization of the sintered magnet of this example of a comparison, the main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) higher than a grain boundary phase was not observed. [0071] Coarse grinding of the ingot alloy J which has the principal component presentation of example of comparison 4 table 7, and the ingot alloy K was carried out like the example 1 except having used, respectively. 81.8kg of coarse powder of Alloy J and 18.2kg of coarse powder of Alloy K were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg mixed coarse powder. When the presentation of mixed coarse powder was analyzed, principal components were Nd:6.00%, B:0.97%, Nb:0.65%, aluminum:0.10%, Co:2.00%, Ga:0.08%, Cu:0.10%, and Remainder: Fe in weight percent, and impurities were O:0.15%, N:0.02%, and C:0.02%. : 19.14%, Pr: 5.34%, Dy .

[0072]

[Table 7]

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
J	22.29	6.21	2.00	0.97	0.80	0.10	2.00	0.08	0.10	残部
K	5.03	1.47	24.00	0.97	-	0.10	2.00	0.08	0.10	残部

[0073] Pulverizing (mean particle diameter of 4.1 micrometers), slurring, and

shaping among a field were performed like the example 1 except having used this coarse powder. the acquired Plastic solid -- about -- the inside of the vacuum of  $5 \times 10^{-1}$  Torr -- 200 degrees C -- 1 hour -- heating -- deoiling -- subsequently -- about -- it cooled to the room temperature after 2-hour sintering in the vacuum of  $2 \times 10^{-5}$  Torr at each temperature in a 1060-1130-degree C temperature requirement. After performing heat treatment of 900 degree-Cx 2 hours, and 500 degree-Cx 1 hour to each obtained sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the sintering mold permanent magnet of the example of a comparison by the blending method was obtained. The result of having measured magnetic properties in 20 degrees C is shown in drawing 3 . When sintering temperature was 1100 degrees C so that clearly from drawing 3 , (BH) max of iHc of Br of 12.1kG(s) and 25.4kOe and 35.1MGOe(s) was obtained, when sintering temperature was 1110 degrees C, (BH) max of iHc of Br of 12.1kG(s) and 25.2kOe and 35.0MGOe(s) was obtained, and Br and (BH) max were low.

[0074] When the presentation of the sintered magnet of this example of a comparison was analyzed, principal components were Nd:19.14%, Pr:0.97%, Nb:0.65%, aluminum:0.10%, Co:2.00%, Ga:0.08%, Cu:0.10%, and Remainder: Fe in weight percent, and impurities were O:0.17%, N:0.06%, and C:0.06%. : 5.34%, Dy : 6.00%, B : It is thought that the thing with low Br of the sintered magnet of this example of a comparison and (BH) max is because the normal grain growth at the time of sintering of the main phase crystal grain was controlled since Nb content is as high as 0.65%.

[0075] Coarse grinding of the ingot alloy L which has the principal component presentation of example 4 table 8, and the ingot alloy M was carried out like the example 1 except having used, respectively. 90.0kg of coarse powder of Alloy L and 10.0kg of coarse powder of Alloy H were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg mixed coarse powder. When the presentation of mixed coarse powder was analyzed, a principal component is Nd at weight percent. : 22.83%, Pr: 6.37%, Dy : 1.30%, B:1.05%, Mo:0.13%, and

aluminum:0.10%, it was Remainder Fe and impurities were O:0.15%, N:0.01%, and C:0.02%.

[0076]

[Table 8]

合金	組成 (重量%)						
	Nd	Pr	Dy	B	Mo	Al	Fe
L	23.85	6.65	—	1.05	0.15	0.10	殘部
M	13.68	3.82	13.00	1.05	—	0.10	殘部

[0077] Pulverizing (mean particle diameter of 4.0 micrometers), slurring, and shaping among a field were performed like the example 1 except having used this mixed coarse powder. the acquired Plastic solid -- about -- the inside of the vacuum of  $5 \times 10^{-1}$  Torr -- 200 degrees C -- 1 hour -- heating -- deoiling -- succeedingly -- about -- it cooled to the after [ 2 hour sintering ] room temperature in the vacuum of  $2 \times 10^{-5}$  Torr at each temperature in a 1050-1100-degree C temperature requirement. After performing heat treatment of 900 degree-Cx 2 hours, and 550 degree-Cx 1 hour to each obtained sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was obtained. As a result of measuring magnetic properties at 20 degrees C, when sintering temperature was 1060-1090 degrees C, magnetic properties desirable as a permanent magnet were acquired. When especially sintering temperature was 1070 degrees C, (BH) max of iHc of Br of 13.9kG(s) and 15.5kOe and 46.5MGOe(s) was obtained, when sintering temperature was 1080 degrees C, (BH) max of iHc of Br of 14.0kG(s) and 15.3kOe and 47.2MGOe(s) was obtained, and Br and (BH) max were high.

[0078] When the presentation of a typical permanent magnet was analyzed among said permanent magnets, principal component presentations were Nd0.13%, aluminum:0.10%, and Remainder: Fe in weight percent, and impurities were O:0.18%, N:0.06%, and C:0.08%. : 22.83%, Pr: 6.37%, Dy : 1.30%, B: 1.05%, Mo :

[0079] The concentration of the heavy-rare-earth element (Dy) in the inside

(almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase was measured like [ organization / of said permanent magnet produced on the sintering temperature of 1070 degrees C, and 1080-degree C conditions / cross-section ] the below-mentioned example 7. Consequently, it turned out that it consists of the first main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, the second main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase, and the third main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase. [0080] Coarse grinding of the ingot alloy N which has the principal component presentation of example 5 table 9, and the ingot alloy O was carried out like the example 1 except having used, respectively. 80.0kg of coarse powder of Alloy N and 20.0kg of coarse powder of Alloy O were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg coarse powder. When the presentation of mixed coarse powder was analyzed, principal components were Nd:0.20%, aluminum:0.1%, Co:2.5%, Cu:0.15%, Ga:0.15%, and Remainder: Fe in weight percent, and impurities were O:0.15%, N:0.02%, and C:0.02%. : 26.2%, Dy : 5.8%, B: 0.95%, Nb

[0081]

[Table 9]

合金	組成 (重量%)								
	Nd	Dy	B	Nb	Al	Co	Cu	Ga	Fe
N	29.0	3.0	0.95	—	0.10	2.50	0.15	0.15	殘部
O	15.0	17.0	0.95	1.00	0.10	2.50	0.15	0.15	殘部

[0082] The oxygen density carried out jet mill grinding in nitrogen-gas-atmosphere mind 500 ppm or less (volume ratio), and used mixed coarse powder as fines with a mean particle diameter of 4.2 micrometers. Dry type compression molding of these fines was carried out on condition that magnetic-field-strength 10kOe and moulding pressure 1.5 ton/cm<sup>2</sup>. The acquired Plastic solid was cooled to the after [ 2 hour sintering ] room temperature in the vacuum of



abbreviation  $3 \times 10^{-5}$  Torr at each temperature in a 1040-1100-degree C temperature requirement.

[0083] After performing heat treatment of 900 degree-Cx 3 hours, and 480 degree-Cx 1 hour to each obtained sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was obtained. When magnetic properties were measured at 20 degrees C, and sintering temperature was made into 1050-1090 degrees C, magnetic properties desirable as a permanent magnet were acquired. When especially sintering temperature was 1070 degrees C, (BH) max of iHc of Br of 12.5kG(s) and 24.5kOe and 37.5MGOe(s) was obtained, when sintering temperature was 1080 degrees C, (BH) max of iHc of Br of 12.5kG(s) and 24.2kOe and 37.4MGOe(s) was obtained, and Br and (BH) max were high. When said permanent magnet was analyzed, principal components were Nd0.20%, aluminum:0.1%, Co:2.5%, Cu:0.15%, Ga:0.15%, and Remainder: Fe in weight percent, and impurities were O:0.38%, N:0.03%, and C:0.05%. : 26.2%, Dy : 5.8%, B: 0.95%, Nb :

[0084] The concentration of the heavy-rare-earth element (Dy) in the inside (almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase was measured like [ organization / of said sintered magnet / whose sintering temperature is 1070 degrees C and 1080 degrees C / cross-section ] the below-mentioned example 7. Consequently, it turned out that it consists of the first main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, the second main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase, and the third main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase. [0085] Coarse grinding of the ingot alloy P which has the principal component presentation of example 6 table 10, and the ingot alloy Q was carried out like the example 1 except \*\*\*\*\*. 90.0kg of coarse powder of Alloy P and 10.0kg of coarse

powder of Alloy Q were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg mixed coarse powder. When the presentation of mixed coarse powder was analyzed, principal components were Nd:0.18%, aluminum:0.05%, Ga:0.17%, and Remainder: Fe in weight percent, and impurities were O:0.15%, N:0.01%, and C:0.01%. : 20.6%, Pr: 8.8%, Dy : 2.6%, B: 1.06%, W

[0086]

[Table 10]

合金	組成 (重量%)							
	Nd	Pr	Dy	B	W	Al	Ga	Fe
P	21.70	9.30	1.00	1.08	0.20	0.05	0.17	殘部
Q	10.50	4.50	17.00	1.06	—	0.05	0.17	殘部

[0087] The oxygen density carried out jet mill grinding in nitrogen-gas-atmosphere mind 500 ppm or less (volume ratio), and used mixed coarse powder as fines with a mean particle diameter of 4.2 micrometers. Dry type compression molding of these fines was carried out on condition that magnetic-field-strength 10kOe and moulding pressure 1.5 ton/cm<sup>2</sup>. The acquired Plastic solid was cooled to the room temperature after 2-hour sintering in the vacuum of abbreviation 3x10<sup>-5</sup> Torr at each temperature in a 1040-1100-degree C temperature requirement.

[0088] After performing heat treatment of 900 degree-Cx 3 hours, and 550 degree-Cx 1 hour to each obtained sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature, and the R-T-B system sintering mold permanent magnet was obtained. When magnetic properties were measured at 20 degrees C, and sintering temperature was made into 1050-1090 degrees C, magnetic properties desirable as a permanent magnet were acquired. When especially sintering temperature was 1070 degrees C, (BH) max of iHc of Br of 13.2kG(s) and 19.5kOe and 41.8MGoe(s) was obtained, when sintering temperature was 1080 degrees C, (BH) max of iHc of Br of 13.2kG(s) and 19.3kOe and 41.7MGoe(s) was obtained, and Br and (BH) max were high.

[0089] When the presentation of a typical permanent magnet was analyzed

among said permanent magnets, it was Nd:0.18%, aluminum:0.05%, Ga:0.17%, and Remainder: Fe in weight percent, and impurities were O:0.50%, N:0.02%, and C:0.06%. : 20.6%, Pr: 8.8%, Dy : 2.6%, B: 1.06%, W

[0090] The concentration of the heavy-rare-earth element (Dy) in the inside (almost core) of the main phase crystal grain (R2T14B) and a grain boundary phase was measured like [ organization / of said permanent magnet produced on the sintering temperature of 1070 degrees C, and 1080-degree C conditions / cross-section ] the below-mentioned example 7. Consequently, it turned out that it consists of the first main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) more expensive the R2T14B type main phase crystal grain than a grain boundary phase, the second main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) lower than a grain boundary phase, and the third main phase crystal grain with the concentration of a heavy-rare-earth element (Dy) almost equal to a grain boundary phase. [0091] Coarse grinding of the ingot alloy R which has the principal component presentation of example 7 table 11, and the ingot alloy S was carried out like the example 1 except having used respectively. 90.0kg of coarse powder of Alloy R and 10.0kg of coarse powder of Alloy S were thrown into the V shaped rotary mixer, and it mixed, and considered as 100kg mixed coarse powder. Remainder: When the presentation of mixed coarse powder was analyzed, in weight percent, Nd1.03%, aluminum:0.08%, Co:2.00%, Ga:0.08%, and Cu:0.1%, the principal component was Fe and were O:0.14%, N:0.02%, and C:0.02%. : 21.38%, Pr: 7.12%, Dy : 1.50%, B :

[0092]

[Table 11]

合金	組成(%)								
	Nd	Pr	Dy	B	Al	Co	Ga	Cu	Fe
R	22.50	7.50	-	1.03	0.08	2.00	0.08	0.10	残部
S	11.25	3.75	15.00	1.03	0.08	2.00	0.08	0.10	残部

[0093] The oxygen density carried out jet mill grinding in nitrogen-gas-atmosphere mind 10 ppm or less (volume ratio), and used mixed coarse powder

as fines with a mean particle diameter of 4.2 micrometers. The obtained fines were collected in nitrogen-gas-atmosphere mind in direct mineral oil (the Idemitsu Kosan make, trade name: Idemitsu super sol PA-30), without making atmospheric air touched. The obtained slurry was pressed by magnetic-field-strength 10kOe and moulding pressure 1.0 ton/cm<sup>2</sup>. In the vacuum of abbreviation 5x10<sup>-1</sup> Torr, the acquired Plastic solid was heated for 1 hour, and was deoiled at 200 degrees C, and it cooled to the room temperature after 2-hour sintering in the vacuum of abbreviation 3x10<sup>-5</sup> Torr at each temperature in a 1040-1100-degree C temperature requirement.

[0094] After performing heat treatment of 900 degree-Cx 2 hours, and 480 degree-Cx 1 hour to each sintered compact once each in an inert gas ambient atmosphere, it cooled to the room temperature and the R-T-B system sintering mold permanent magnet was produced. When magnetic properties were measured at 20 degrees C, and sintering temperature was 1060-1090 degrees C, magnetic properties desirable as a permanent magnet were acquired. When especially sintering temperature was 1070 degrees C, (BH) max of iHc of Br of 13.9kG(s) and 15kOe and 46.5MGoe(s) was obtained, when sintering temperature was 1080 degrees C, (BH) max of iHc of Br of 14.0kG(s) and 14.8kOe and 47.2MGoe(s) was obtained, and Br and (BH) max were high.

[0095] When the typical sintered magnet was analyzed among said sintered magnets, principal components were Nd1.50%, B:1.03%, aluminum:0.08%, Co:2.00%, Ga:0.08%, Cu:0.1%, and Remainder: Fe in weight percent, and impurities were O:0.16%, N:0.06%, and C:0.06%. : 21.38%, Pr: 7.12%, Dy :

[0096] The typical cross-section organization of a sintered magnet was analyzed on condition that the following using EPMA (the product made from Electron Probe Micro-Analyzer; JEOL, form JXA-8800) among said sintered magnets.

Acceleration voltage: The rectangle range, [0097] which are 0.12micrometer and area:0.12micrometerx400 point =48micrometer of field analysis : 15kV, sample absorption current: Abbreviation 4x10<sup>-8</sup>A, X-ray incorporation time amount per one analysis point (gate time) : 10msec, the number of analysis points: Both the

direction of length (X) and the direction of width (Y) are spacing of each analysis point of 400 points, X, and the direction of Y. By extracting a beam to the minimum spot and irradiating it on the above-mentioned conditions, concentration distribution of Dy, Nd, and Pr was measured. The analysing crystal used for analysis of Dy, Nd, and Pr was a high sensitivity type \*\*\*\*-ized lithium (LiF). The crystalline structure of the R-T-B system sintering mold permanent magnet of this example is roughly shown in drawing 4 . The crystalline structure has the R2T14B type main phase crystal grain 1 and the grain boundary phase 2, and a black field shows 3 important 2' of a grain boundary phase. Moreover, the concentration distribution of Dy in the crystalline structure of drawing 4 is shown in drawing 5 , concentration distribution of Nd is shown in drawing 6 R> 6 (a), and concentration distribution of Pr is shown in drawing 6 (b). Although distribution of Nd, Dy, and Pr was substantially accepted with 3 importance in the grain boundary phase so that clearly from drawing 5 , drawing 6 R> 6 (a), and (b), Nd, Dy, and Pr are not necessarily distributed only over 3 importance, and this is because a grain boundary phase is very thin, so there are very few amounts of distribution of Nd, Dy, and Pr except 3 importance.

[0098] In drawing 4 , R (Nd, Dy, Pr) rich phase forms 3 importance of a grain-boundary phase. Drawing 6 (a) and (b) show that Nd and Pr exist in homotopic mostly. Moreover, although Dy existed also in the field of the almost same grain boundary phase as Nd and Pr from drawing 5 , drawing 6 (a), and (b), it turned out that it may exist in high concentration also in the part in the R2T14B type main phase crystal grain which separated 1.0 micrometers or more from the grain boundary (core part).

[0099] These observation results showed that there was a pattern of three kinds of Dy concentration distribution about Dy concentration distribution which reaches the core in the main phase crystal grain from a grain boundary phase. By the first pattern, the core part in the main phase crystal grain has Dy concentration higher than a grain boundary phase. By the second pattern, Dy concentration of a grain boundary phase is high, and Dy concentration of the

core part in the main phase crystal grain is low. Dy concentration distribution from a grain boundary phase to [ the third pattern ] the core of the main phase crystal grain is almost uniform. In drawing 5 , the number of the third main phase crystal grain which the number of the first main phase crystal grain with higher core part [ in the main phase crystal grain ] Dy concentration is six, and the number of the second main phase crystal grain with low Dy concentration is 15, and has [ phase / grain boundary ] Dy concentration almost equal to a grain boundary phase from a grain boundary phase was 19. in addition, drawing 5 , drawing 6 (a), and (b) -- setting -- Dy and Nd -- and -- When evaluating concentration distribution of Pr, the effect of the void formed in the creation time of the sample for microscope observation was taken into consideration. Moreover, it is not necessary to pass over drawing 4 , drawing 5 and drawing 6 (a), and (b) to an example of a cross-section organization but, and they need to average the data for which the cross-section organization in many visual fields was asked for determining concentration distribution of Dy. Thus, the R-T-B system sintering mold permanent magnet of this invention has characteristic Dy concentration distribution in the main phase crystal grain and a grain boundary phase.

[0100] The particle size distribution of the main phase crystal grain of a typical thing is shown in drawing 7 among said permanent magnets. The axis of abscissa of drawing 7 shows the size range of the main phase crystal grain, for example, "9-10 micrometers" means that the size range of the main phase crystal grain is "9 micrometers or more less than 10 micrometers." Using the optical microscope (form UFX-II, NIKON Make), the particle size of the main phase crystal grain took the photograph (one 1000 times the scale factor of this) of the cross section of the arbitration of a permanent magnet, and carried out the image processing of this cross-section photograph with the image-processing software (Image Pro.Plus (DOS/V)) by the PURANE TRON company. The cross-section configuration of each \*\*\*\* crystal grain was further assumed to be a circle by having set to Si area of each \*\*\*\* crystal grain of the arbitration measured by the image processing, and each diameter  $d_i$  of \*\*\*\* crystal grain was defined as

one (4x Si/pi) half. The rate of distribution of an axis of ordinate (%) shows the ratio  $[(TN/T) \times 100\%]$  of the number TN of the main phase crystal grain in each particle-size within the limits to the total T of the main phase crystal grain in the measured visual field.

[0101] In the permanent magnet of this invention, the rate of distribution of the main phase crystal grain with a particle size of less than 2 micrometers was 0%, and the rate of distribution of the main phase crystal grain 16 micrometers or more was 5.8% so that clearly from drawing 7 . As a result of inquiring furthermore, the rate of distribution of the main phase crystal grain with a particle size of less than 2 micrometers was less than 5%, and when the rate of distribution of the main phase crystal grain 16 micrometers or more was 10% or less, it turned out that magnetic properties desirable as a permanent magnet are realizable. further -- the rate of distribution of the main phase crystal grain with a particle size of less than 2 micrometers -- 3% or less -- and the rate of distribution of the main phase crystal grain 16 micrometers or more is 8% or less -- more -- desirable -- the rate of distribution of the main phase crystal grain with a particle size of less than 2 micrometers -- 0% -- and it turned out that it is desirable that the rate of distribution of the main phase crystal grain 16 micrometers or more is especially 6% or less. In addition, said main phase particle size distribution can be realized even when Nb content is 0.01 - 0.6%.

[0102] Coarse grinding was carried out like the example 7 except having used the ingot alloy T which has the principal component presentation of example of comparison 5 table 12. When the presentation of coarse powder was analyzed, principal components were Nd:1.50%, B:1.03%, Nb:0.70%, aluminum:0.08%, Co:2.00%, Ga:0.08%, Cu:0.1%, and Remainder: Fe in weight percent, and impurities were O:0.15%, N:0.01%, and C:0.02%. : 21.38%, Pr: 7.12%, Dy

[0103]

[Table 12]

合金	組成(%)									
	Nd	Pr	Dy	B	Nb	Al	Co	Ga	Cu	Fe
T	21.38	7.12	1.50	1.03	0.70	0.08	2.00	0.08	0.10	殘部

[0104] Using this coarse powder, like the example 7, pulverizing (mean particle diameter of 4.1 micrometers), slurring, shaping among a field, deoiling, sintering, and heat treatment were performed, and the sintering mold permanent magnet of the example of a comparison by the single method was obtained. When the presentation of this sintered magnet was analyzed, principal components were Nd1.03%, Nb:0.70%, aluminum:0.08%, Co:2.00%, Ga:0.08%, Cu:0.1%, and Remainder: Fe in weight percent, and impurities were O:0.17%, N:0.05%, and C:0.07%. : 21.38%, Pr:7.12%, Dy : 1.50%, B :

[0105] Although the level of iHc was as high as 16kOe(s) order as a result of measuring magnetic properties in 20 degrees C, Br(s) were 13.5 or less kGs and less than [ (BH) max44.0MGoe ], and were low compared with the example 7.

[0106] Drawing 8 shows the cross-section organization of this sintered magnet roughly. Three in the crystalline structure shows a void and other numbers are the same as drawing 4 . It was checked from drawing 8 Dy concentration distribution almost uniform from a grain boundary phase to the core of the main phase crystal grain and that Dy concentration of a grain boundary phase is high, and two patterns of the distribution with almost low Dy concentration of a core in the main phase crystal grain exist. The number of the main phase crystal grain which has the almost same Dy concentration distribution as a grain boundary phase was 31, and that of the main phase crystal grain with Dy concentration lower than a grain boundary phase was 15. However, the distribution with almost high Dy concentration of a core in the main phase crystal grain was not observed from a grain boundary phase.

[0107] The result of having evaluated the diameter distribution of the main phase crystal grain of the sintered magnet of this example of a comparison like the example 7 is shown in drawing 9 . In this sintered magnet, the rate of distribution of the with a 1-micrometer or more particle size [ less than 2 micrometer ] main phase crystal grain is 12.5%, and, on the whole, the diameter distribution of the main phase crystal grain has shifted to the diameter side of a granule greatly compared with distribution of drawing 7 , and the main phase crystal grain has



not carried out grain growth enough so that clearly from drawing 9  $R > 9$ . For this reason, it is judged compared with an example 7 that Br and (BH) max are low.

[0108] Although the above-mentioned example indicated the case where a heavy-rare-earth element was Dy, also in Tb or Ho, the R-T-B system sintering mold permanent magnet which has the main phase crystal grain with the concentration of Tb or Ho higher than a grain boundary phase in a core part almost like the case of Dy, and has high Br and high (BH) max like the above-mentioned example can be obtained.

[0109] In the above-mentioned example, with the same R content, and two sorts of R-T-B system alloy powder whose principal components of other correspond except that only ratios which constitute R element, such as Dy and Nd, differ, With the same R content, or and by mixing using two sorts of R-T-B system alloy powder whose principal components of other correspond except refractory metal elements (Nb etc.) having permuted ratios, such as Dy, Nd, etc. which constitute R element, and a part of Fe The R-T-B system sintering mold permanent magnet which has the main phase crystal grain which has characteristic Dy concentration distribution, and has the suitable diameter distribution of the main phase crystal grain for the high application of Br and (BH) max was able to be obtained to stability. In this invention, three or more sorts of R-T-B system alloy powder may be used as said R-T-B system alloy powder. Moreover, mixing of these R-T-B system alloy powder may be performed in a fines phase.

[0110] Various kinds of surface preparation (nickel plating, electrodeposited epoxy resin coating, etc.) can be used for the R-T-B system sintering mold permanent magnet of the above-mentioned example suitable for \*\*\*\*\* and various kinds of applications (actuators, such as a voice coil motor or CD pickup, or rotating machine).

[0111]

[Effect of the Invention] As explained in full detail above the R-T-B system sintering mold permanent magnet of this invention The first R2T14B type main phase crystal grain with heavy-rare-earth element concentration (Dy etc.) more

expensive the R2T14B type main phase crystal grain than a grain boundary phase, Since it consists of the second R2T14B type main phase crystal grain with heavy-rare-earth element concentration (Dy etc.) lower than a grain boundary phase, and the third R2T14B type main phase crystal grain with heavy-rare-earth element concentration (Dy etc.) almost equal to a grain boundary phase While having the R-T-B system sintering mold permanent magnet obtained by the single method, and iHc high to the same extent, it has higher Br and higher (BH) max. Therefore, the R-T-B system sintering mold permanent magnet of this invention can be used suitable for the application as which high Br and high (BH) max are required.

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[Translation done.]

\* NOTICES \*

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- 1.This document has been translated by computer. So the translation may not reflect the original precisely.
- 2.\*\*\*\* shows the word which can not be translated.
- 3.In the drawings, any words are not translated.

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## DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] It is the graph which shows the relation between sintering temperature and magnetic properties (Br, iHc) about the R-T-B system sintering mold permanent magnet of an example 1 and the example 1 of a comparison.

[Drawing 2] It is the graph which shows the relation between sintering temperature and magnetic properties (Br, iHc) about the R-T-B system sintering

mold permanent magnet of an example 2 and the example 2 of a comparison.

[Drawing 3] It is the graph which shows the relation between sintering temperature and magnetic properties ( $B_r$ ,  $iH_c$ ) about the R-T-B system sintering mold permanent magnet of an example 3 and the examples 3 and 4 of a comparison.

[Drawing 4] It is the schematic diagram showing the crystalline structure of the R-T-B system sintering mold permanent magnet of an example 7.

[Drawing 5] It is the EPMA photograph in which the concentration distribution of Dy in the crystalline structure of the R-T-B system sintering mold permanent magnet of an example 7 is shown.

[Drawing 6] Concentration distribution of the heavy-rare-earth element in the crystalline structure of the R-T-B system sintering mold permanent magnet of an example 7 is shown, (a) is an EPMA photograph in which the concentration distribution of Nd in the crystalline structure is shown, and (d) is an EPMA photograph in which concentration distribution of Pr in the crystalline structure is shown.

[Drawing 7] It is the graph which shows the particle size distribution of the main phase crystal grain in the R-T-B system sintering mold permanent magnet of an example 7.

[Drawing 8] It is the schematic diagram showing the crystalline structure of the R-T-B system sintering mold permanent magnet of the example 5 of a comparison.

[Drawing 9] It is the graph which shows the particle size distribution of the main phase crystal grain in the R-T-B system sintering mold permanent magnet of the example 5 of a comparison.

[Description of Notations]

1 ... The R2T14B type main phase crystal grain

2 ... Grain boundary phase

2' ... Triple point

3 ... Void

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[Translation done.]

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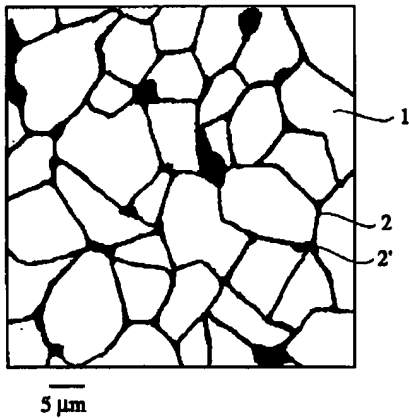
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**DRAWINGS**

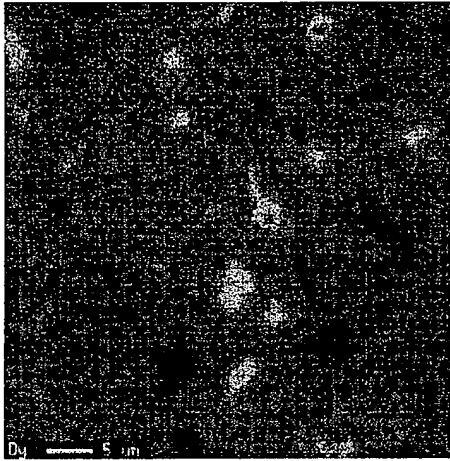
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[Drawing 4]

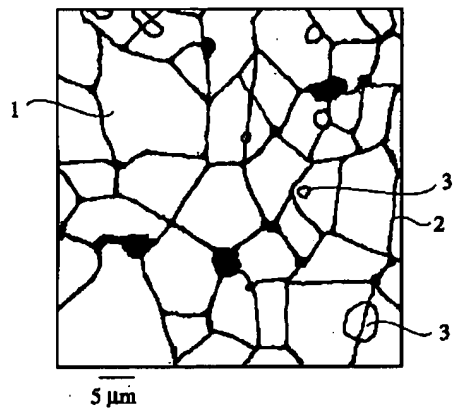
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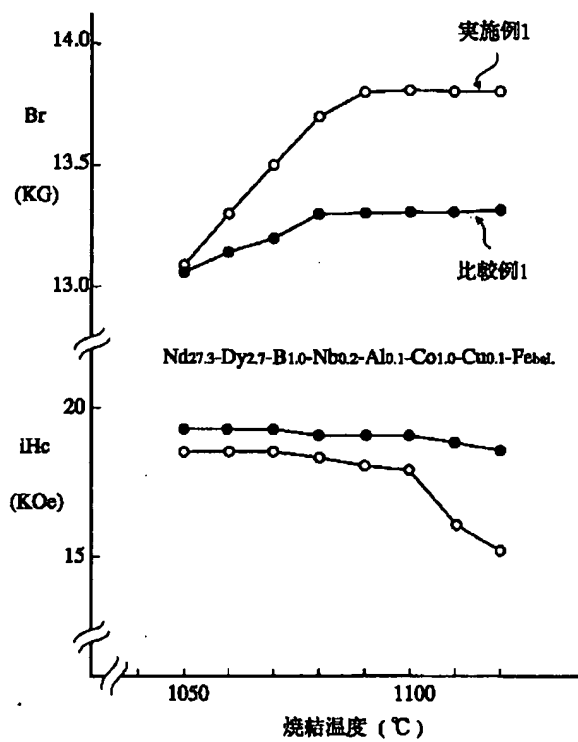
[Drawing 5]



[Drawing 8]

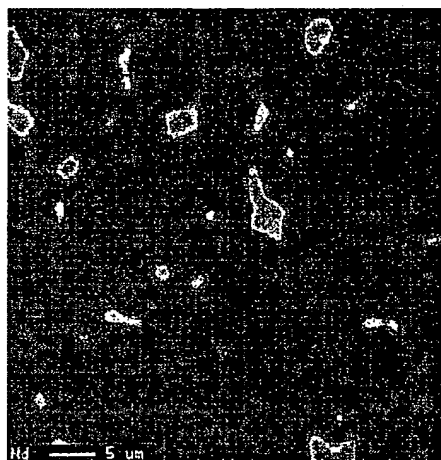


[Drawing 1]

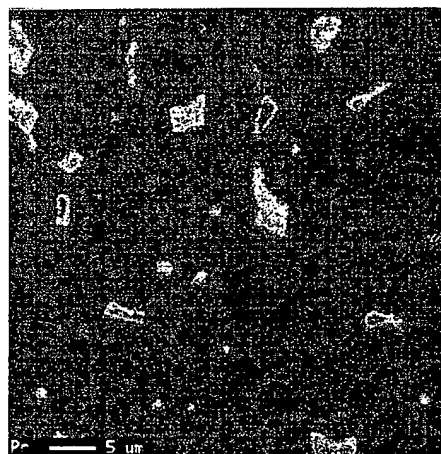




( a )



( b )



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[Translation done.]